

LA-UR-18-21915

Approved for public release; distribution is unlimited.

Title: Advancements in Fission Modeling for Nuclear Applications

Author(s): Jaffke, Patrick John

Intended for: Job Interview

Issued: 2018-03-08

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



Advancements in Fission Modeling for Nuclear Applications

Patrick Jaffke
T-2 Postdoc, Los Alamos National Lab

UNCLASSIFIED

Slide 1

Collaborators

- P. Talou¹, I. Stetcu¹, T. Kawano¹, P. Möller¹, A. Sierk¹
- B. Byerly², L. Tandon², A. Hayes¹, G. Jungman¹
- S. Okumura³, S. Chiba³
- M. Devlin⁴, N. Fotiades⁴
- N. Schunck⁵, M. Mumpower¹

¹ Theoretical Division, Los Alamos National Lab

² Chemistry Division, Los Alamos National Lab

³ Tokyo Institute of Technology

⁴ Physics Division, Los Alamos National Lab

⁵ Nuclear Data and Theory Group, Lawrence Livermore National Lab

UNCLASSIFIED

Outline

1. Historical overview of nuclear fission
 - Current status of models and theory
2. Importance of fission modeling
 - Fundamental science, nonproliferation, criticality, heating, etc.
3. Applications:
 - Expanding Pu suite for multiplicity/criticality
 - Couple theory models to provide reasonable predictions
 - Creating new diagnostic tools with simplified depletion
 - New solutions in the very-low-burnup regime
4. Conclusions and outlook

UNCLASSIFIED

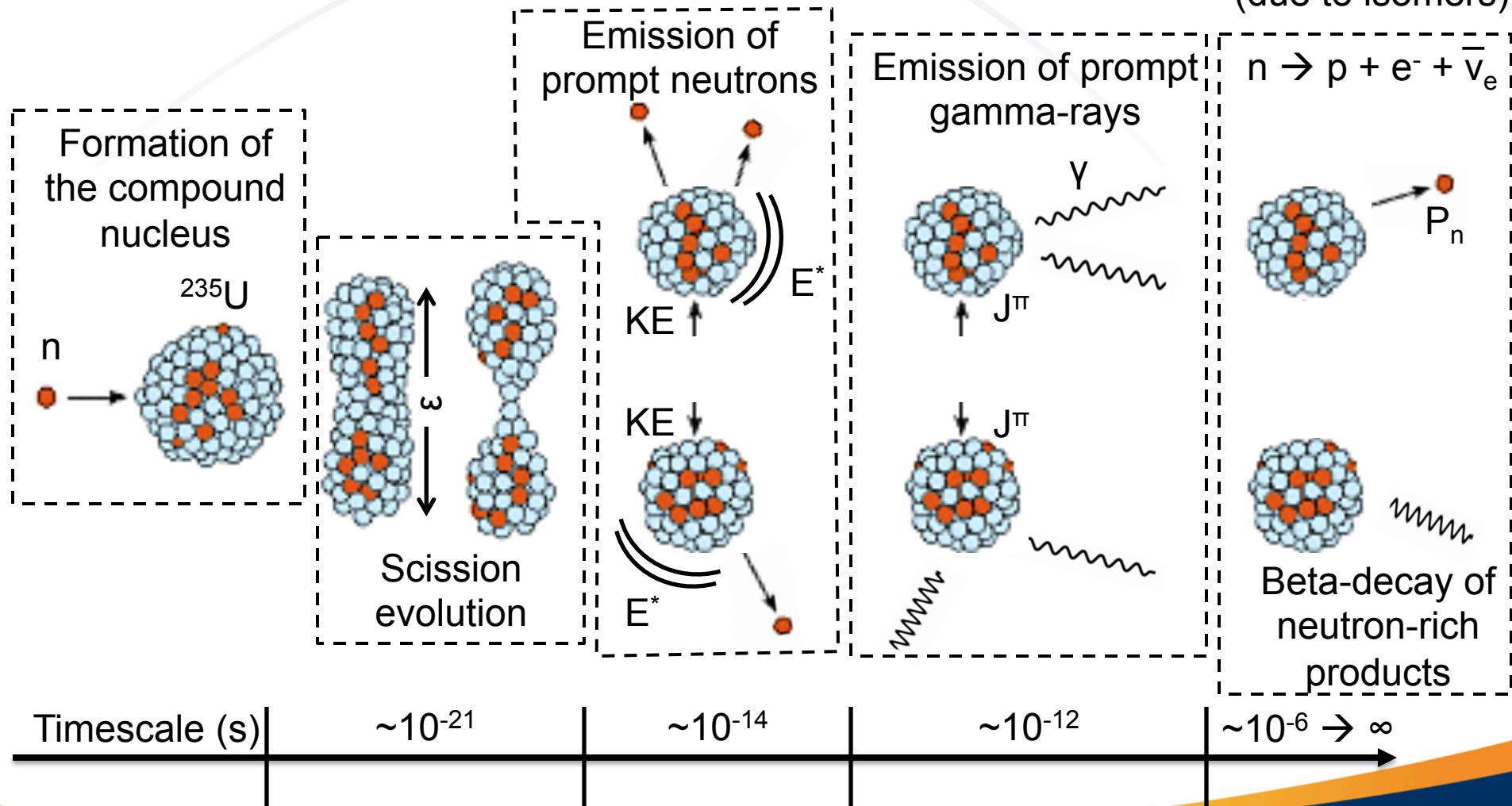
What is the fission process?

$$Q = TXE + TKE$$

$$Y_{\text{pre}}(A, Z)$$

$$Y_{\text{post}}(A, Z)$$

Delayed neutrons
and gamma-rays
(due to isomers)



UNCLASSIFIED

Outline

1. Historical overview of nuclear fission
 - Current status of models and theory
2. Importance of fission modeling
 - Fundamental science, nonproliferation, criticality, heating, etc.
3. Applications:
 - Expanding Pu suite for multiplicity/criticality
 - Couple theory models to provide reasonable predictions
 - Creating new diagnostic tools with simplified depletion
 - New solutions in the very-low-burnup regime
4. Conclusions and outlook

UNCLASSIFIED

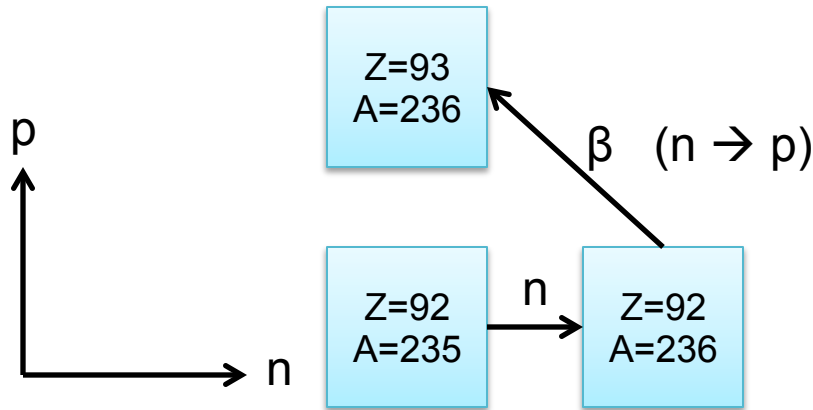
History of nuclear fission

1. An accidental finding

- 1934: Fermi bombards Uranium with neutrons believing he has produced heavier elements ($Z=93$)



Enrico Fermi



Ida Noddack

amount to produce near neighboring elements. When heavy nuclei are bombarded by neutrons, it is conceivable that the nucleus breaks up into several large fragments, which would of course be isotopes of known elements but would not be neighbors of the irradiated element.

Über das Element 93

Dr.-Ing. Ida Noddack (1934)

UNCLASSIFIED

History of nuclear fission

2. Fission confirmed!



Otto Hahn



Fritz Strassmann



- 1938: Hahn and Strassmann identify Barium after n
→ Uranium

!! Roughly $\frac{1}{2}$ the mass of Uranium !!

Disintegration of Uranium by Neutrons: a
New Type of Nuclear Reaction (1939)

History of nuclear fission

3. Model Development

- 1939: Bohr and Wheeler apply liquid drop model for fission

$$E_B = a_V A - a_S A^{2/3} - a_C Z^2 / A^{1/3} - a_A (N - Z)^2 / A + \delta(A, Z)$$

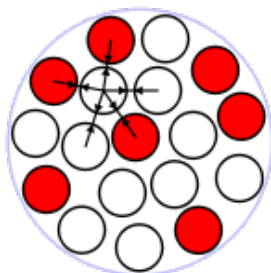
Volume term

Surface correction

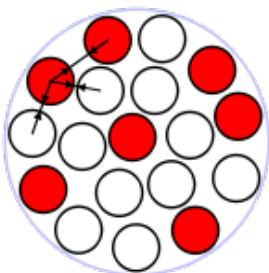
Coulomb interaction

Isospin dependence

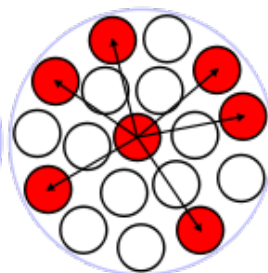
Pairing term



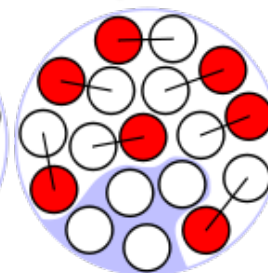
Volume



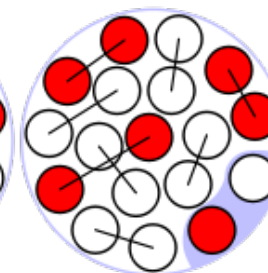
Surface



Coulomb



Asymmetry



Pairing

UNCLASSIFIED

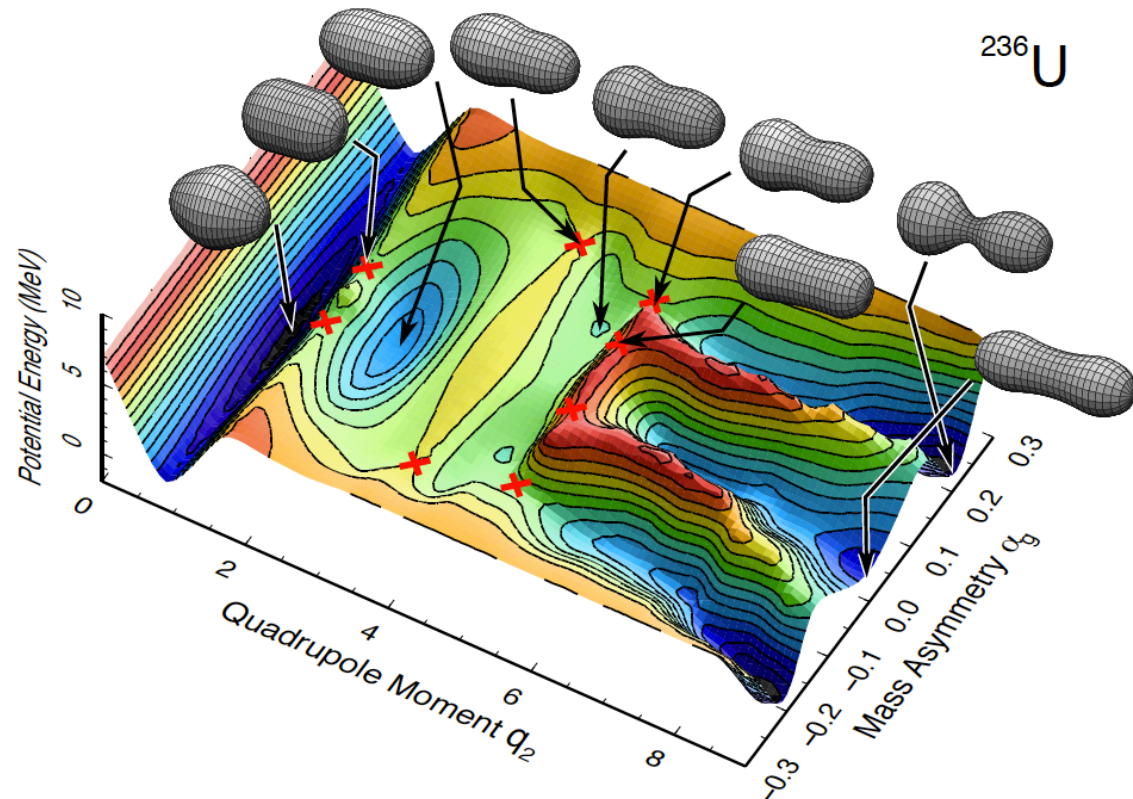
History of nuclear fission

4. Scission Evolution (today)

- Macro-micro: semi-classical (shape + nuclear corrections)

T. Ichikawa *et al.* arXiv:1203.2011v2 (2011)

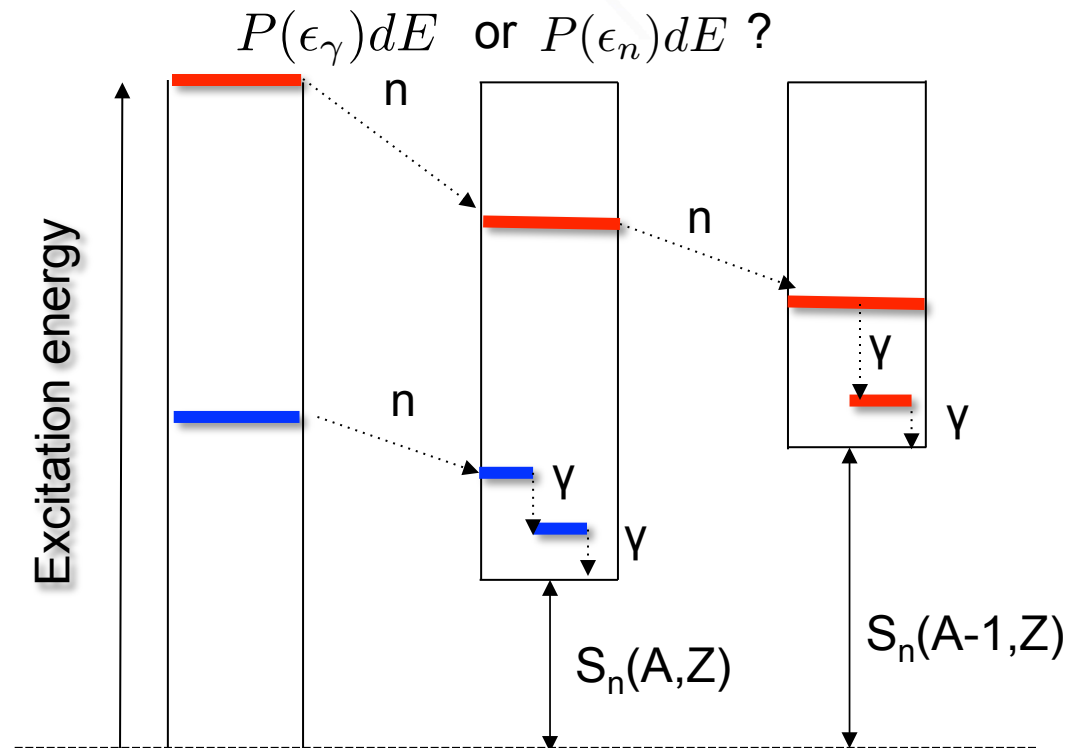
- Just a few model parameters fitted with nuclear masses
- Provides $Y_{\text{pre}}(A, Z)$ for unmeasured reactions!!



History of nuclear fission

5. Fragment De-excitation (today)

- **CGMF**: Monte Carlo implementation of Hauser-Feshbach
- Requires starting distribution of fission fragments:
 - $Y(A, Z, E^*, J^\pi)$
 - A few model parameters
- **Provides prompt neutron and γ -ray emissions!!**



UNCLASSIFIED

Outline

1. Historical overview of nuclear fission
 - Current status of models and theory
2. Importance of fission modeling
 - Fundamental science, nonproliferation, criticality, heating, etc.
3. Applications:
 - Expanding Pu suite for multiplicity/criticality
 - Couple theory models to provide reasonable predictions
 - Creating new diagnostic tools with simplified depletion
 - New solutions in the very-low-burnup regime
4. Conclusions and outlook

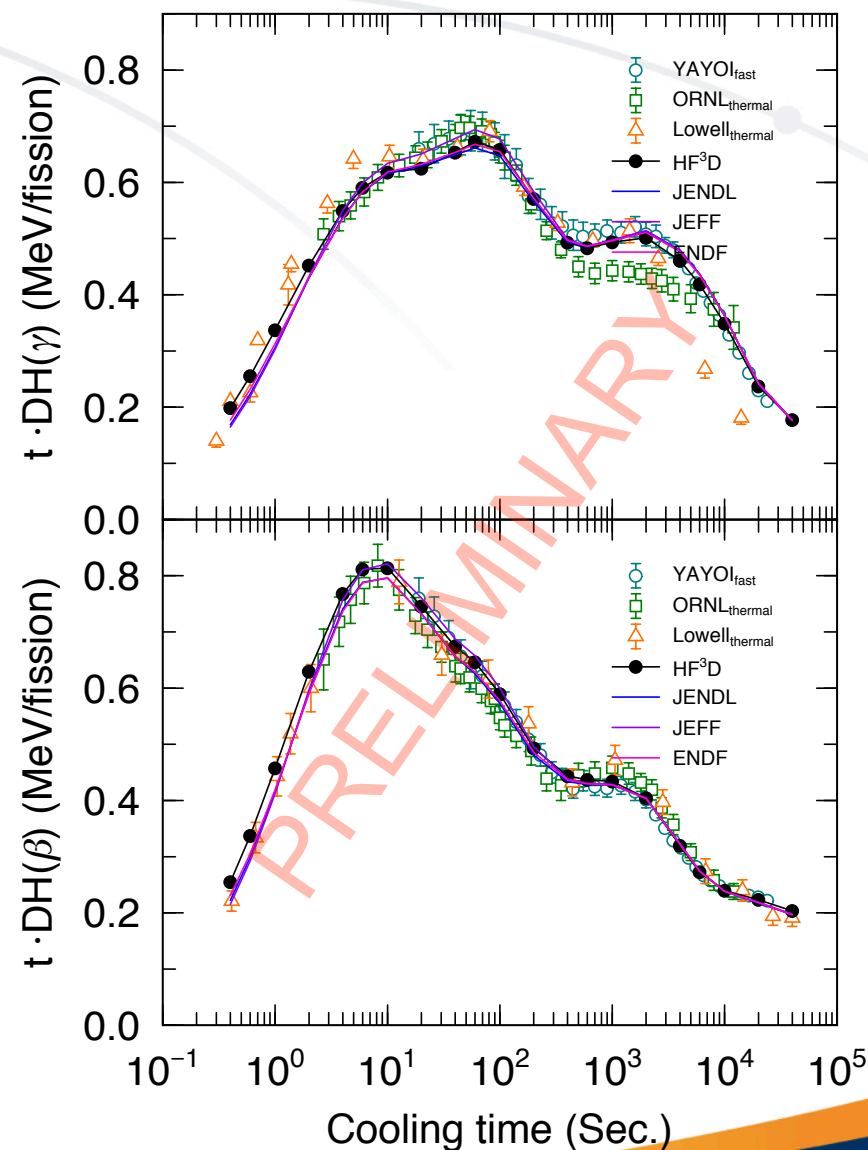
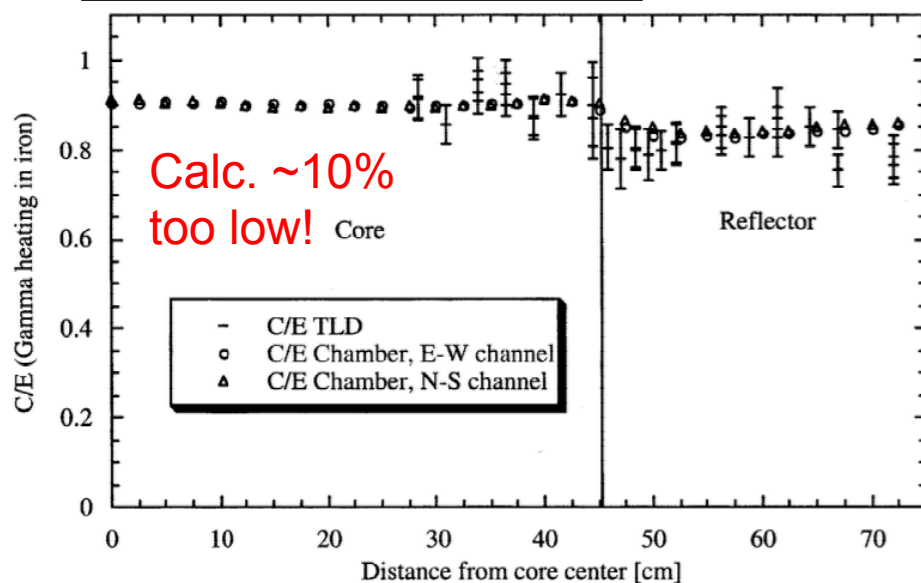
UNCLASSIFIED

Some motivations

1. Next-gen reactor design

- Accurate energy release for new fuels, designs, etc.
- Energy release from fragments, β 's, γ -rays

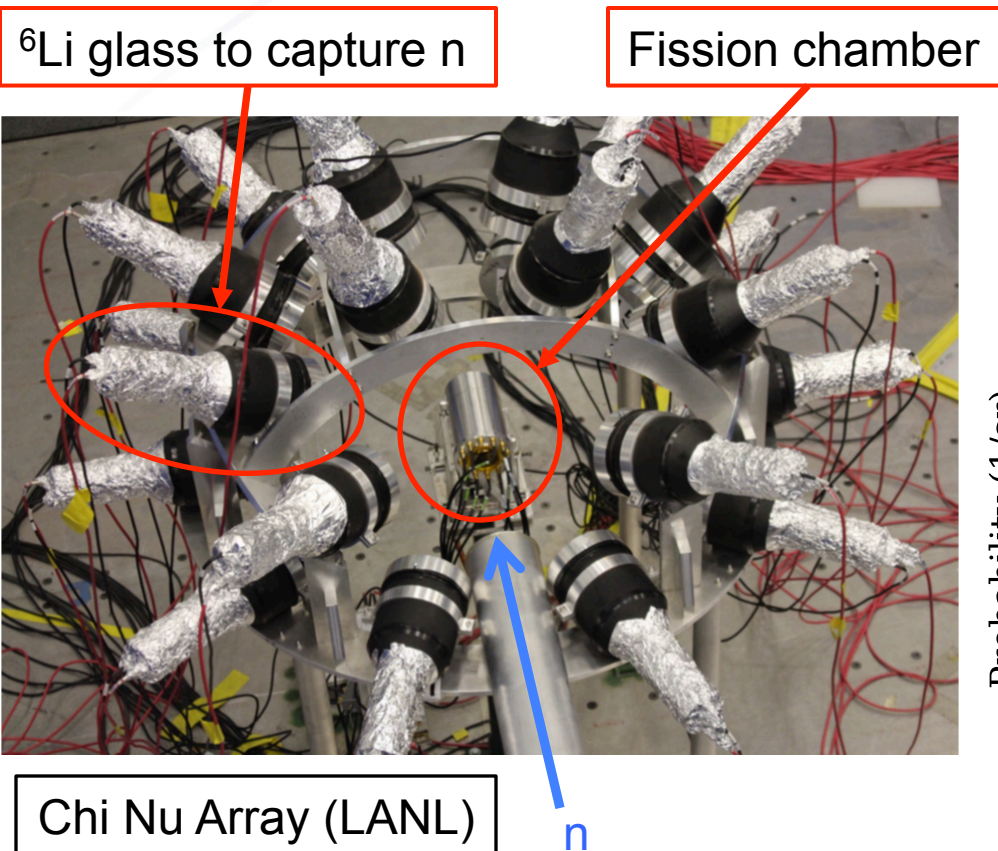
Lüthi, NSE **138** 3 (2001)



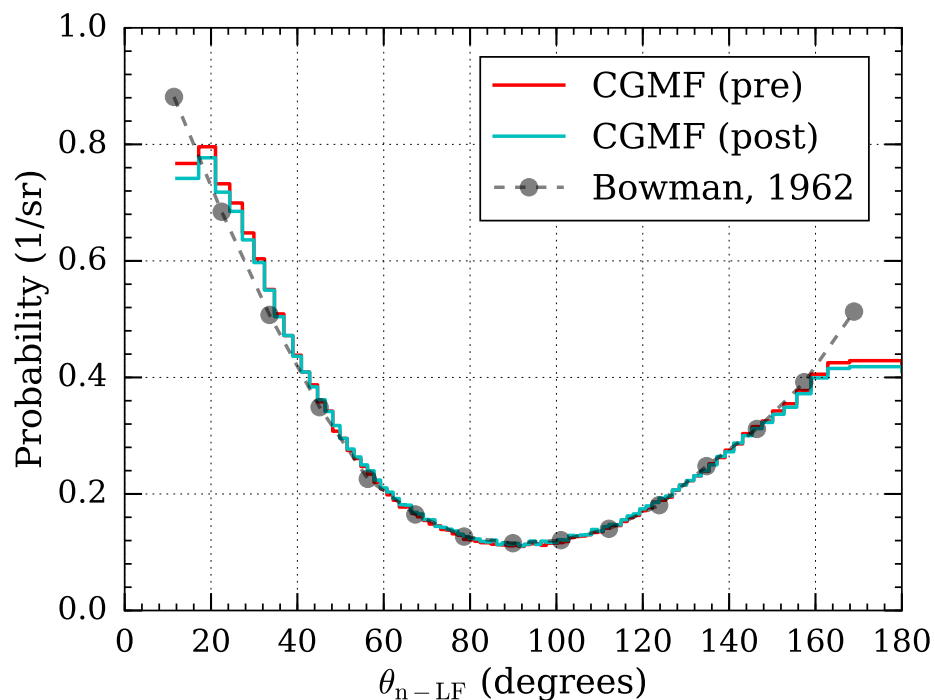
UNCLASSIFIED

Some motivations

2. Verify trends in new correlated experimental data or guide new experimental designs



Neutrons predominantly emitted in direction of fragments (0° or 180°)
!! Direct impact on applications !!



UNCLASSIFIED

Slide 13

Some motivations

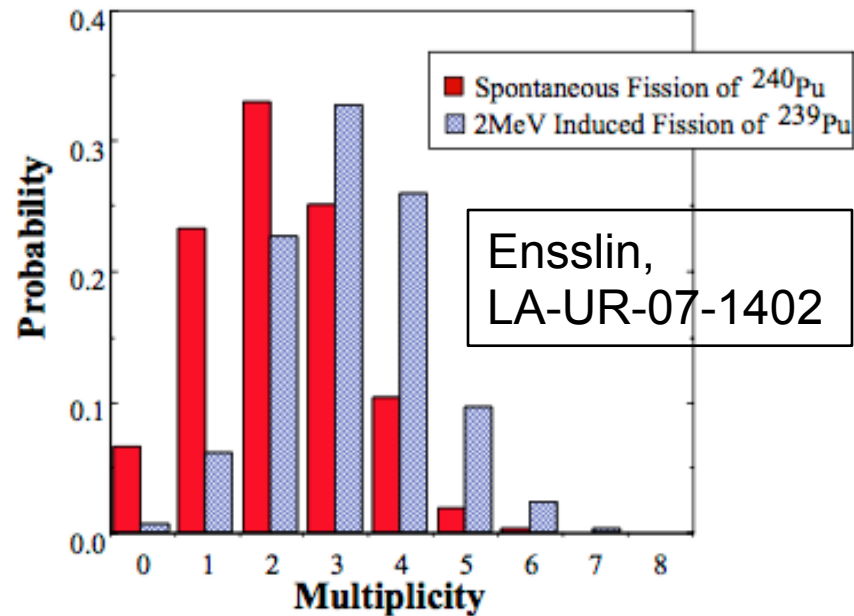
3. Predictions for various applications

- Multiplicity counting for source identification

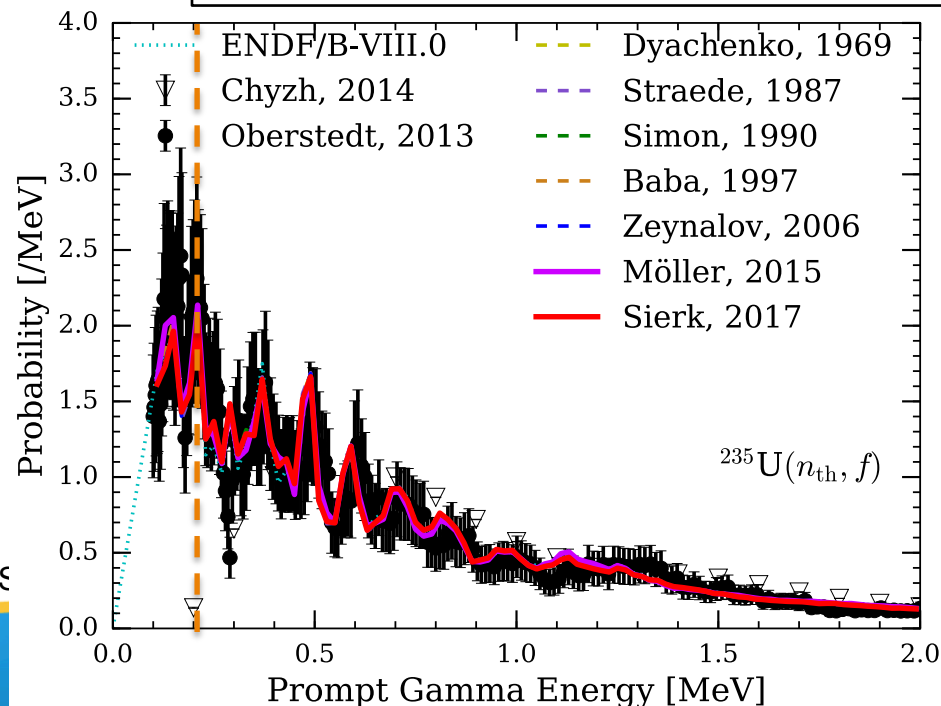
Multiplicity distributions different for **Pu240sf** and **Pu239nf** reactions!

- Identification of isotopes from γ -ray spectroscopy

Fission product abundance related to intensity of specific γ -ray lines!
213 keV line corresponds to ^{100}Zr



PJ et al. arXiv:1712.05511 (2017)



UNCLASS

Outline

1. Historical overview of nuclear fission
 - Current status of models and theory
2. Importance of fission modeling
 - Fundamental science, nonproliferation, criticality, heating, etc.
3. Applications:
 - Expanding Pu suite for multiplicity/criticality
 - Couple theory models to provide reasonable predictions
 - Creating new diagnostic tools with simplified depletion
 - New solutions in the very-low-burnup regime
4. Conclusions and outlook

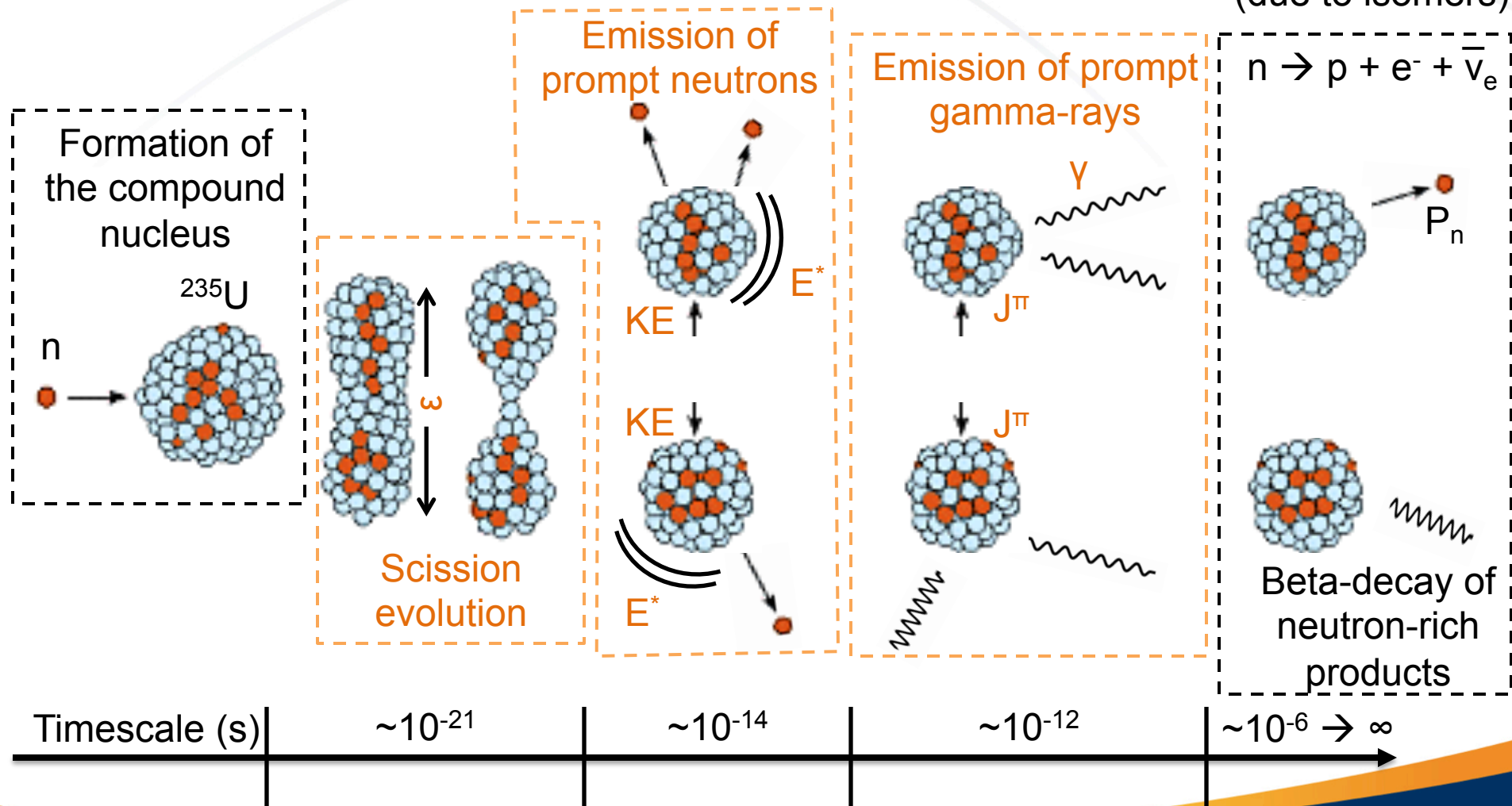
UNCLASSIFIED

What is the fission process?

$Y_{\text{post}}(A, Z)$
Delayed neutrons
and gamma-rays
(due to isomers)

$$Q = TXE + TKE$$

$$Y_{\text{pre}}(A, Z)$$



UNCLASSIFIED

Expanding the plutonium suite

- Available data:

Isotope/Reaction	$\bar{\nu}$ (n/fiss)	P_v	ϵ_n Spectrum
Pu236(sf)	Hicks (1965), Crane (1956)	Hicks (1956)	×
Pu238(sf)	Hicks (1965), Crane (1956)	Hicks (1956)	×
Pu238(n,f)	Jaffey (1970)	×	×
Pu239(n,f)	Frehaut (1980), Khokhlov (1976)	Holden (1988), Boldeman (1985)	Chatillon (2014), Nefedov (1983)
Pu240(sf)	Huanqiao (1984)	Boldeman (1985)	Gerasimenko (2002)
Pu240(n,f)	Khokhlov (1994), Frehaut (1974)	×	Smith (1980)
Pu241(n,f)	Frehaut (1974), Vorobyeva (1974)	Holden (1988), Boldeman (1985)	×
Pu242(sf)	Boldeman (1985)	Boldeman (1985)	Gerasimenko (2002)
Pu242(n,f)	Khokhlov (1994)	×	×

Expanding the plutonium suite

- Objective: provide reasonable estimates for prompt neutron multiplicity, distribution, and energies
 - And ν , P_ν , and ϵ_n depend on incident neutron energy E_n
- Issues:
 - CGMF requires $Y_{\text{pre}}(\mathbf{A}, \mathbf{Z}, \mathbf{E}^*, \mathbf{J}^\pi)$
 - Need some mixture of theory + systematics guidance

Q-value

$$TXE = \overbrace{E_n + B_n + M(A_0, Z_0) - M(A_L, Z_L) - M(A_H, Z_H)}^{\text{Q-value}} - TKE$$

✓ ✓ ✓

✗ ✗

✗

From nuclear data

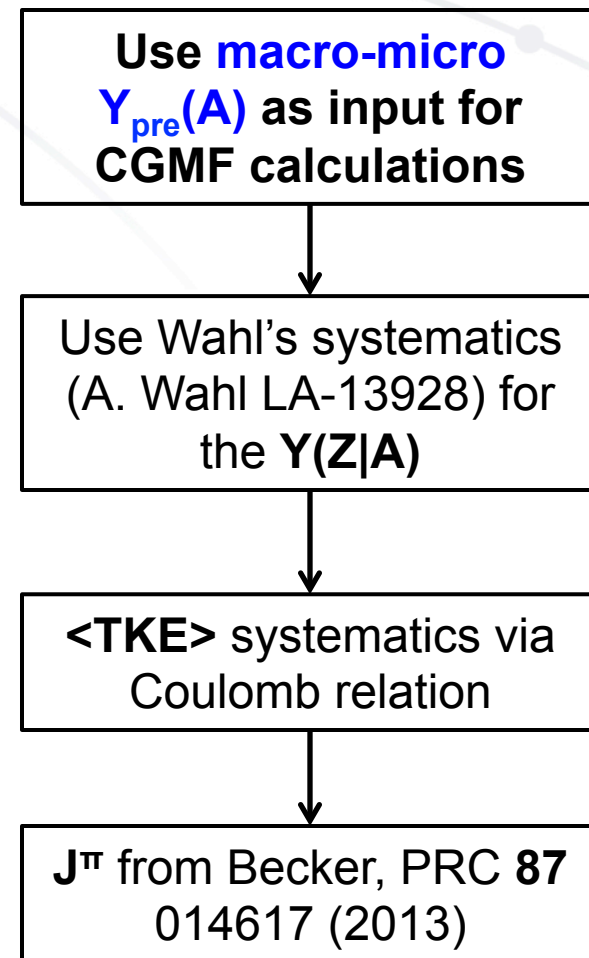
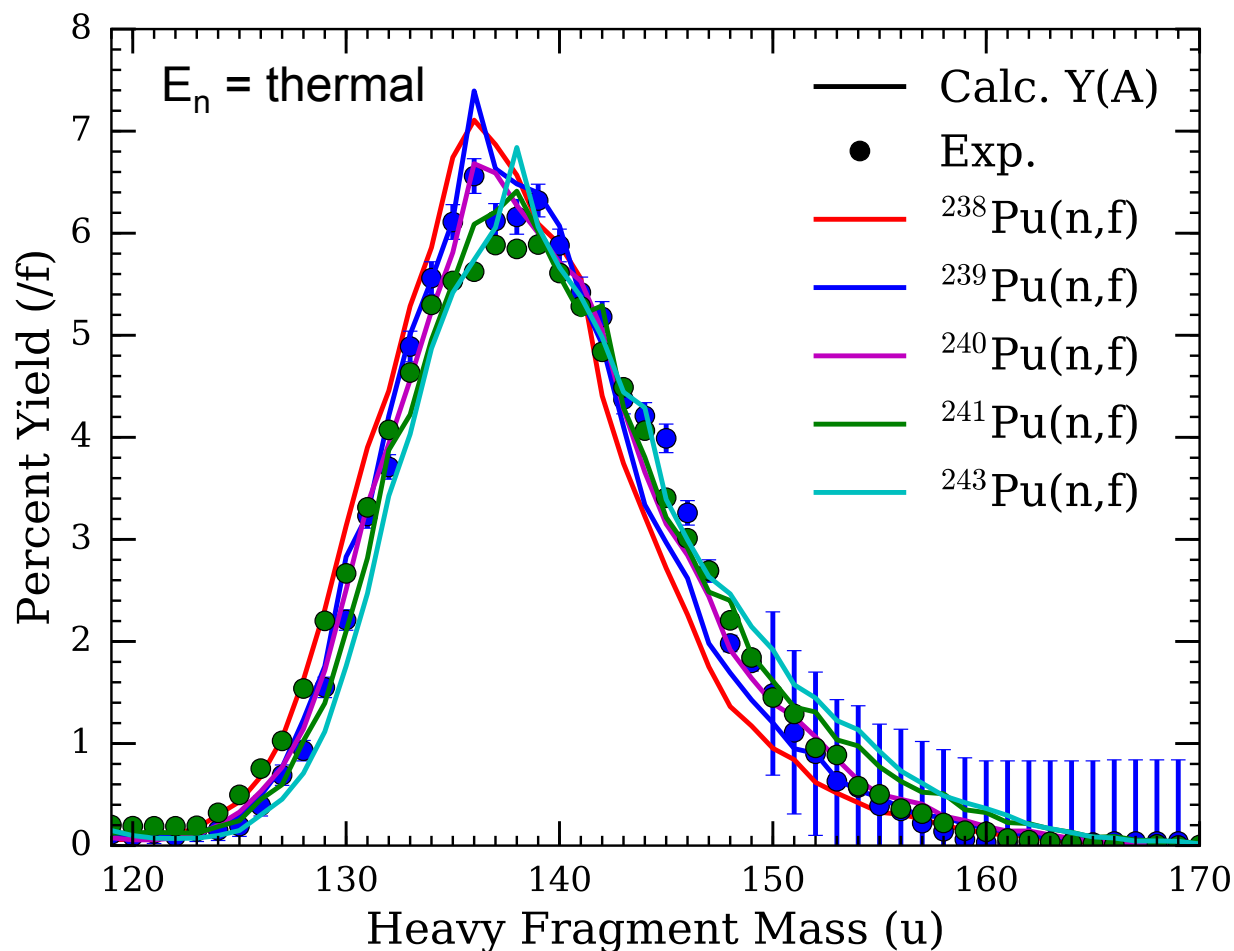
Use theory $Y_{\text{pre}}(\mathbf{A}, \mathbf{Z})$?

Systematics?

UNCLASSIFIED

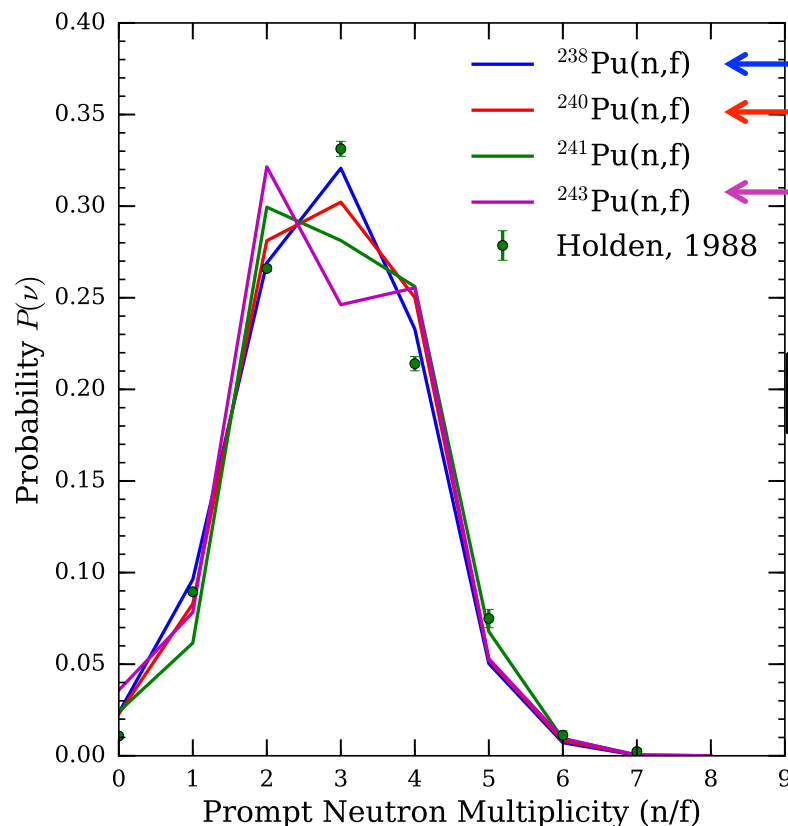
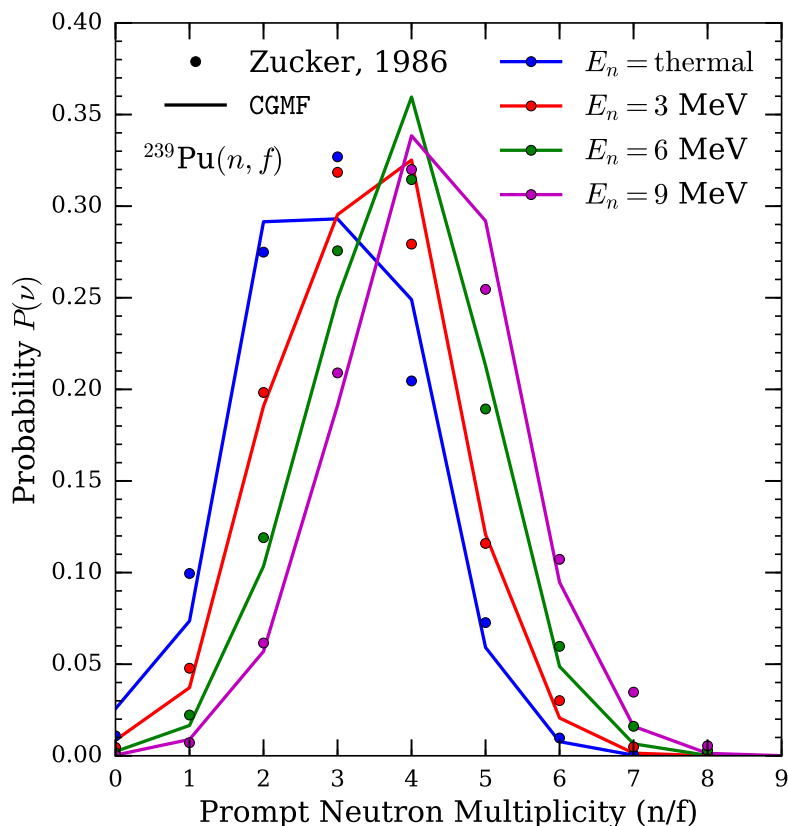
Start with macro-micro $Y_{\text{pre}}(A)$

- Comparing theory with experiment
 - Reasonable agreement with data



Predictions for neutron characteristics

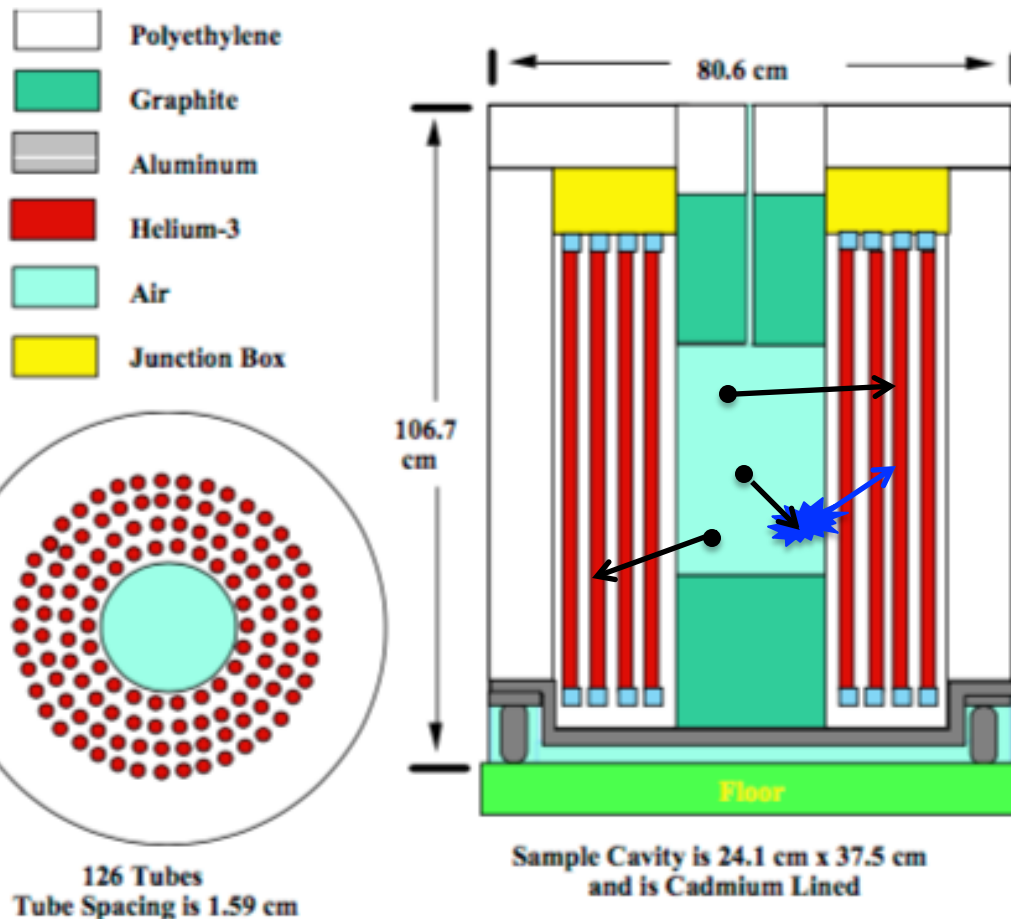
- P_ν shows a similar energy-dependence
 - Shift towards higher neutron multiplicity with increasing E_n
 - Not a lot of data available though...



Predictions!

Neutron multiplicity counting problem

- Count multiplicity of neutrons in a time window
 - Relate singles/doubles/triples to effective ^{240}Pu mass
 - Prompt neutrons from SF can induce fission in rest of Pu!



- Can use calculated P_v for unmeasured fission reactions
- Estimate the multiplication factor

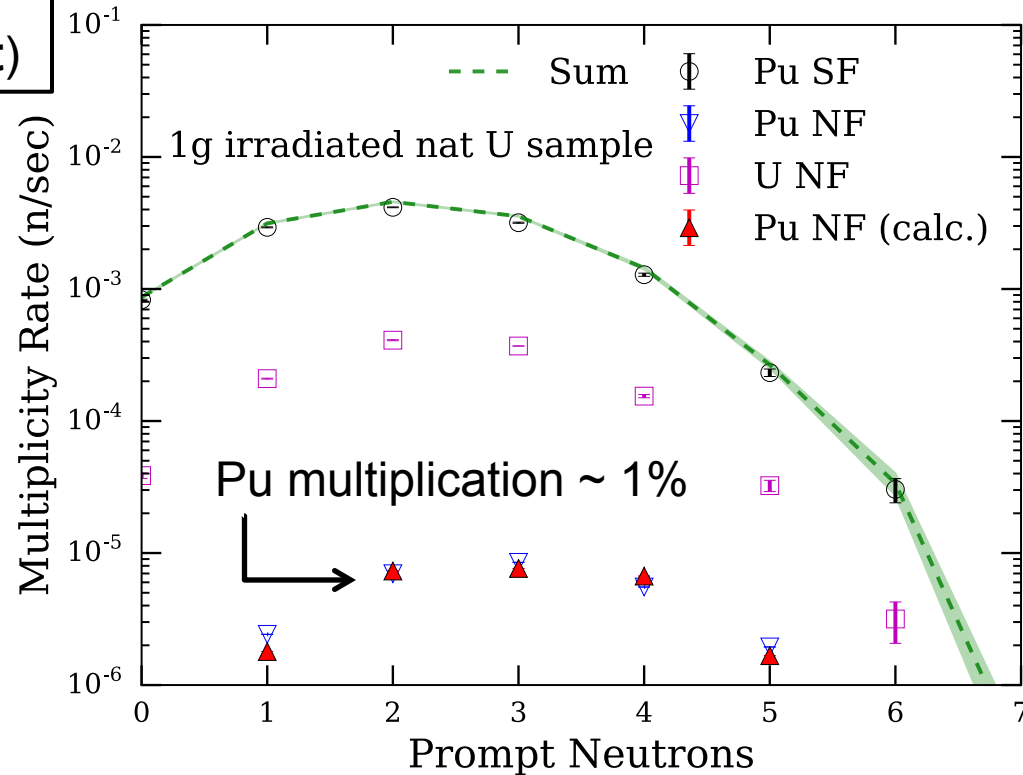
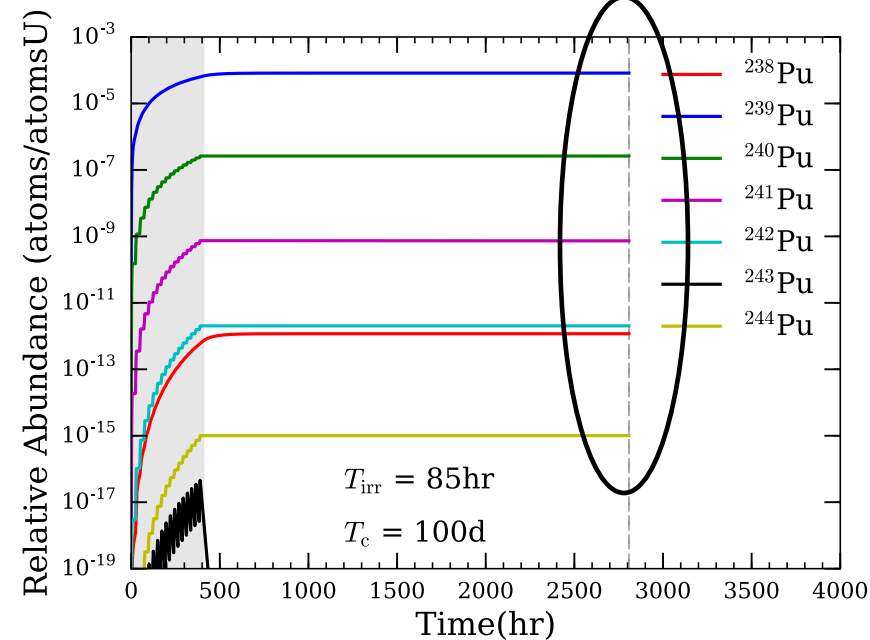
Ensslin, LA-UR-07-1402

Mar. 8, 2018

Neutron multiplicity counting problem

- Get starting isotopics from depletion calculation
 - Determine spontaneous fission P_v rate
 - Calculate the additional P_v from neutron-induced reactions

Starting isotopics favor $^{239}\text{Pu}(n,f)$ which has exp. P_v (calc. P_v have little impact)



UNCLASSIFIED

Slide 22

Outline

1. Historical overview of nuclear fission
 - Current status of models and theory
2. Importance of fission modeling
 - Fundamental science, nonproliferation, criticality, heating, etc.
3. Applications:
 - Expanding Pu suite for multiplicity/criticality
 - Couple theory models to provide reasonable predictions
 - Creating new diagnostic tools with simplified depletion
 - New solutions in the very-low-burnup regime
4. Conclusions and outlook

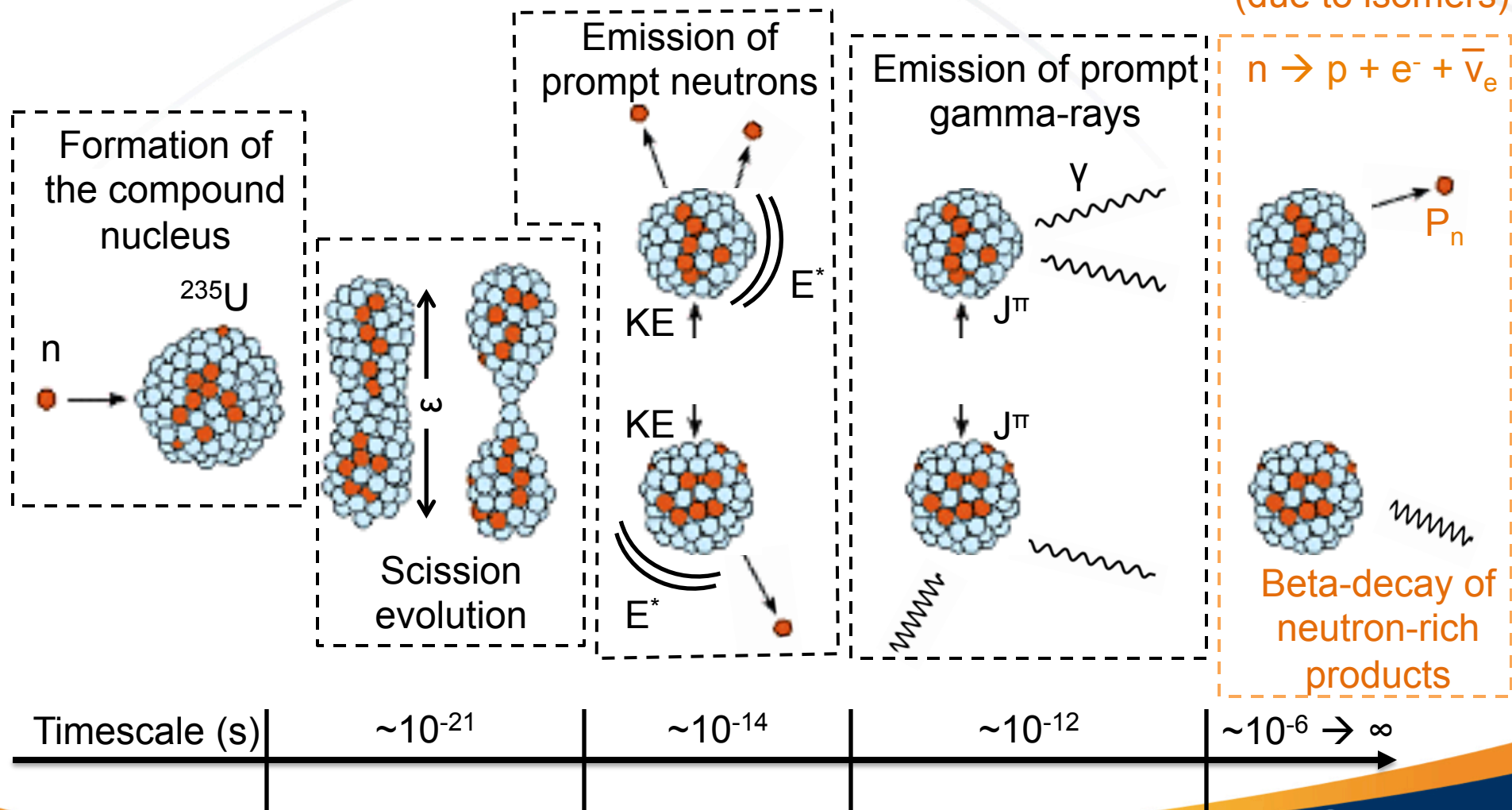
UNCLASSIFIED

What is the fission process?

$$Q = TXE + TKE$$

$$Y_{\text{pre}}(A, Z)$$

$Y_{\text{post}}(A, Z)$
Delayed neutrons
and gamma-rays
(due to isomers)



UNCLASSIFIED

Developing diagnostics for very-low burnup

■ Some basic reactor physics:

^{238}Am 98 M ϵ : 100.00% α : 1.0E-4%	^{239}Am 11.9 H ϵ : 99.99% α : 0.01%	^{240}Am 50.8 H ϵ : 100.00% α : 1.9E-4%	^{241}Am 432.6 Y α : 100.00% SF: 4E-10%	^{242}Am 16.02 H β^- : 82.70% ϵ : 17.30%
^{237}Pu 45.64 D ϵ : 100.00% α : 4.2E-3%	^{238}Pu 87.7 Y α : 100.00% SF: 1.9E-7%	^{239}Pu 24110 Y α : 100.00% SF: 3.E-10%	^{240}Pu 6561 Y α : 100.00% SF: 5.7E-6%	^{241}Pu 14.329 Y β^- : 100.00% α : 2.5E-3%
^{236}Np 153E+3 Y ϵ : 86.30% β^- : 13.50%	^{237}Np 2.144E+6 Y α : 100.00% SF: 2E-10%	^{238}Np 2.117 D β^- : 100.00%	^{239}Np 2.356 D β^- : 100.00%	^{240}Np 61.9 M β^- : 100.00%
^{235}U 7.04E+8 Y 0.7204% α : 100.00% SF: 7.0E-9%	^{236}U 2.342E7 Y α : 100.00% SF: 9.4E-8%	^{237}U 6.75 D β^- : 100.00%	^{238}U 4.468E9 Y 99.2742% α : 100.00% SF: 5.4E-5%	^{239}U 23.45 M β^- : 100.00%

Primary fuel sources:
 LEU (~3%), nat U (0.71%)

UNCLASSIFIED

- **Neutron exposure** gives information about Pu production
- **Cooling time** dates the capabilities
- Depletion analysis uses reaction network to model the buildup of isotopes in a reactor environment

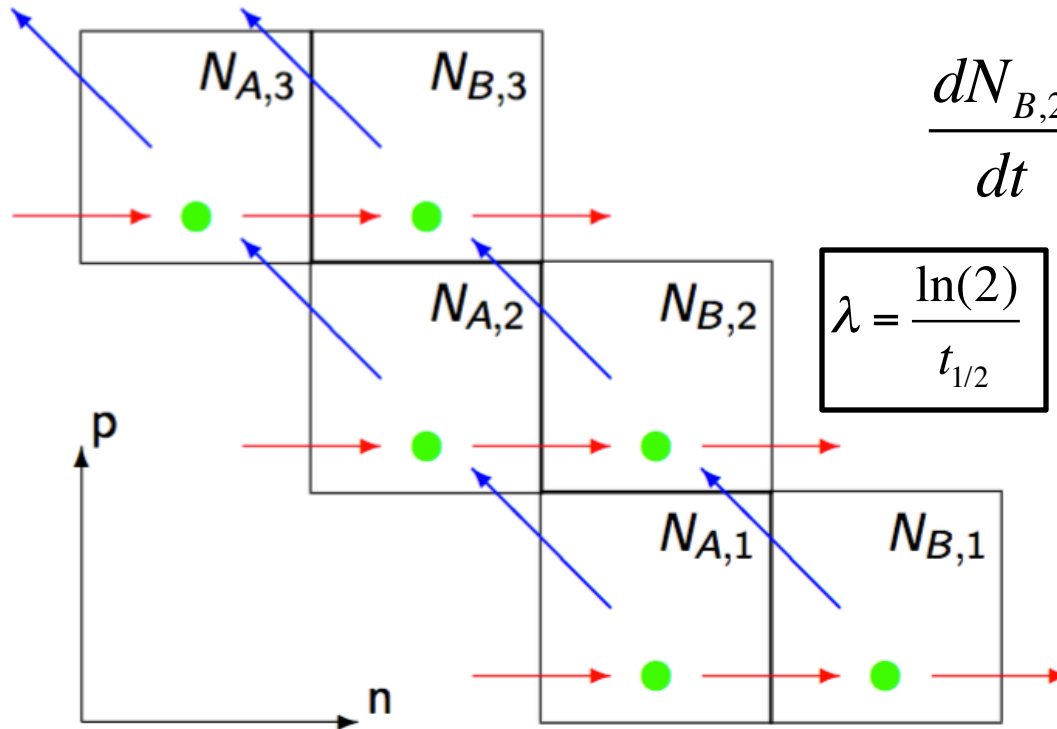
Developing diagnostics for very-low burnup

- Objective: determine the total neutron exposure and the age of reactor samples
 - Neutron exposure Φ_n relates to burnup
 - Cooling time of sample helps date the capabilities
- Issues:
 - Typical methods are not ideal in very-low burnup
 - Not enough $^{241}\text{Pu}/^{241}\text{Am}$ produced for age estimate
 - Not enough ^{134}Cs or ^{154}Eu produced for measurement
 - Graphite Isotope Ratio Method (GIRM) too invasive
 - Additional problems if the samples are very old!

UNCLASSIFIED

Developing diagnostics for very-low burnup

- Procedure: use Bateman equations to develop simplified depletion calculations



$$\lambda = \frac{\ln(2)}{t_{1/2}}$$

Production via fission

$$\frac{dN_{B,2}}{dt} = \vec{Y}_{B,2} \cdot \vec{F}$$

$$\vec{F} = \{F_{U235}, F_{U238}, F_{Pu239}, F_{Pu241}\}$$

Production/Depletion via β -decay

$$+ \lambda_{B,1} N_{B,1} - \lambda_{B,2} N_{B,2}$$

Production/Depletion via n-capture

$$+ \phi_n \sigma_{A,2} N_{A,2} - \phi_n \sigma_{B,2} N_{B,2}$$

- Need decay constants (λ), cross-sections (σ), the fission rate (F), the fission yields (Y), and neutron flux (ϕ_n)

UNCLASSIFIED

Developing diagnostics for very-low burnup

- Example: Neutron exposure Φ_n from Uranium ratio

^{235}U	^{236}U	^{237}U	^{238}U
$\lambda = 1\text{e-}9\text{s}^{-1}$			$\lambda = 2\text{e-}10\text{s}^{-1}$
$\sigma_n = 99\text{b}$			$\sigma_n = 2.7\text{b}$
$\sigma_f = 585\text{b}$	$N = 0$	$N = 0$	$\sigma_f = 2\text{e-}5\text{b}$

^{235}U not produced in fission

$$\frac{dN_{U235}}{dt} = \vec{Y}_{U235} \cdot \vec{F}$$

No ^{235}Pa and $\lambda_{U235} \sim 0$

$$+ \lambda_{Pa235} N_{Pa235} - \lambda_{U235} N_{U235}$$

No ^{234}U

$$+ \phi_n \sigma_{U234} N_{U234} - \phi_n \sigma_{U235} N_{U235}$$

■ ^{235}U

$$\sigma_f \gg \sigma_n$$

$$N_{U235}(t) = N_{U235}^0 e^{-\sigma_{U235} \phi_n t}$$

■ ^{238}U

$$\sigma_n \gg \sigma_f$$

$$\Phi_n = \phi_n t$$

$$N_{U238}(t) = N_{U238}^0 e^{-\sigma_{U238} \phi_n t}$$

$$\varepsilon = \varepsilon_0 e^{-\Phi_n (\sigma_{U235}^T - \sigma_{U238}^T)}$$

UNCLASSIFIED

Developing diagnostics for very-low burnup

- Cooling Time: Ratios of linear fission products

- Linear in Φ_n
- Long half-life w.r.t. T_{irr} and T_C
- Short precursor halflives

- $Z_{Cs137} = Y_{Cs137} + Y_{Xe137} + Y_{I137}$

$$T_C = \frac{1}{\lambda_2 - \lambda_1} \ln \left(\frac{\alpha_{1/2} \lambda_2 \vec{Z}_2 \cdot \langle \vec{\Sigma}_{fiss} \rangle_{\Phi}}{\lambda_1 \vec{Z}_1 \cdot \langle \vec{\Sigma}_{fiss} \rangle_{\Phi}} \right)$$

$$\vec{Z} = \{Z_{U235}, Z_{U238}, Z_{Pu239}, Z_{Pu241}\}$$

└ Cumulative yields

$$\langle \vec{\Sigma}_{fiss} \rangle_{\Phi}$$

Flux-weighted macroscopic fission cross-section

$$\alpha_{1/2} = \frac{\lambda_1 N_1}{\lambda_2 N_2}$$

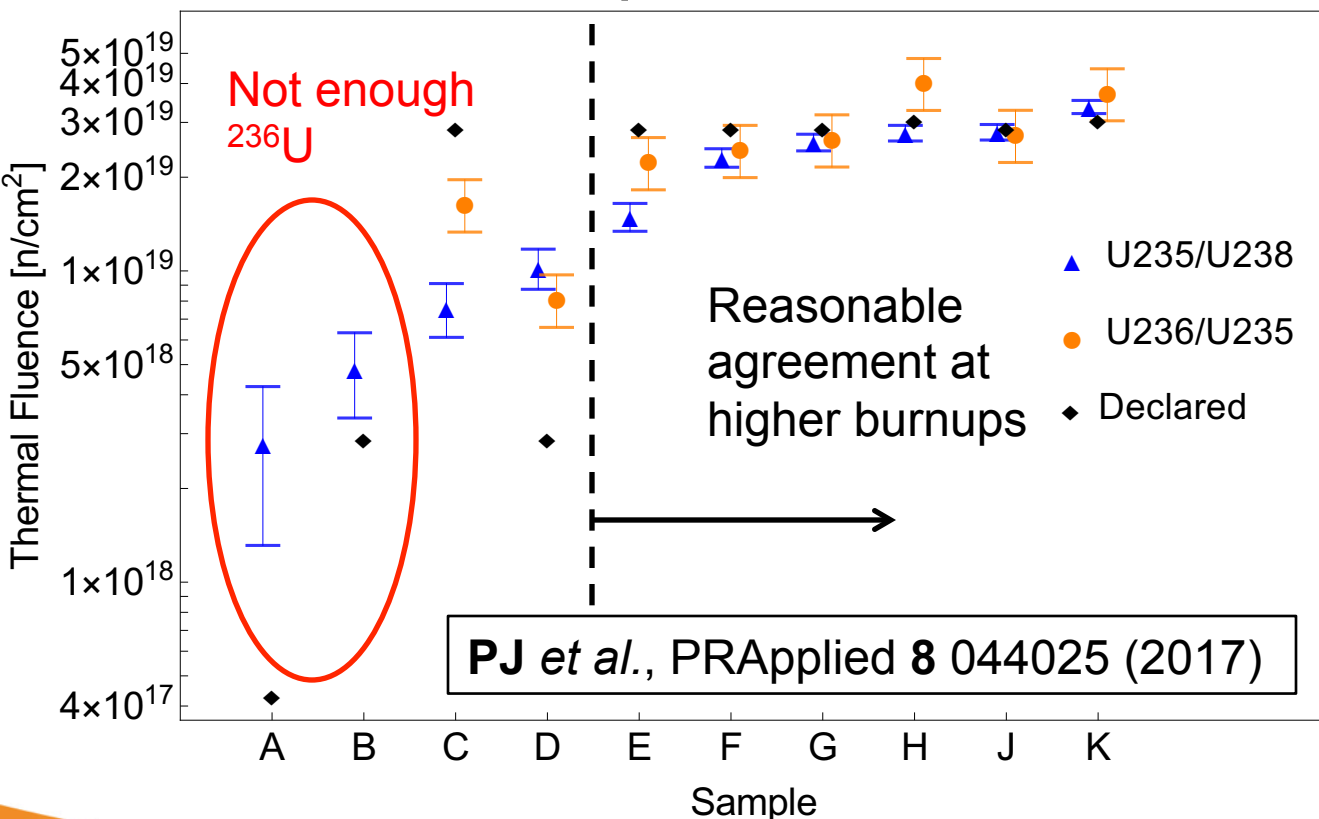
Activity ratio of products 1 & 2

^{137}Cs 30.08 Y β^- : 100.00%	^{138}Cs 33.41 M β^- : 100.00%	^{139}Cs 9.27 M β^- : 100.00%
^{136}Xe >2.4E+21 Y 8.8573% 2 β^-	^{137}Xe 3.818 M β^- : 100.00%	^{138}Xe 14.08 M β^- : 100.00%
^{135}I 6.58 H β^- : 100.00%	^{136}I 83.4 S β^- : 100.00%	^{137}I 24.5 S β^- : 100.00% β^- -n: 7.14%

UNCLASSIFIED

Applying the diagnostics

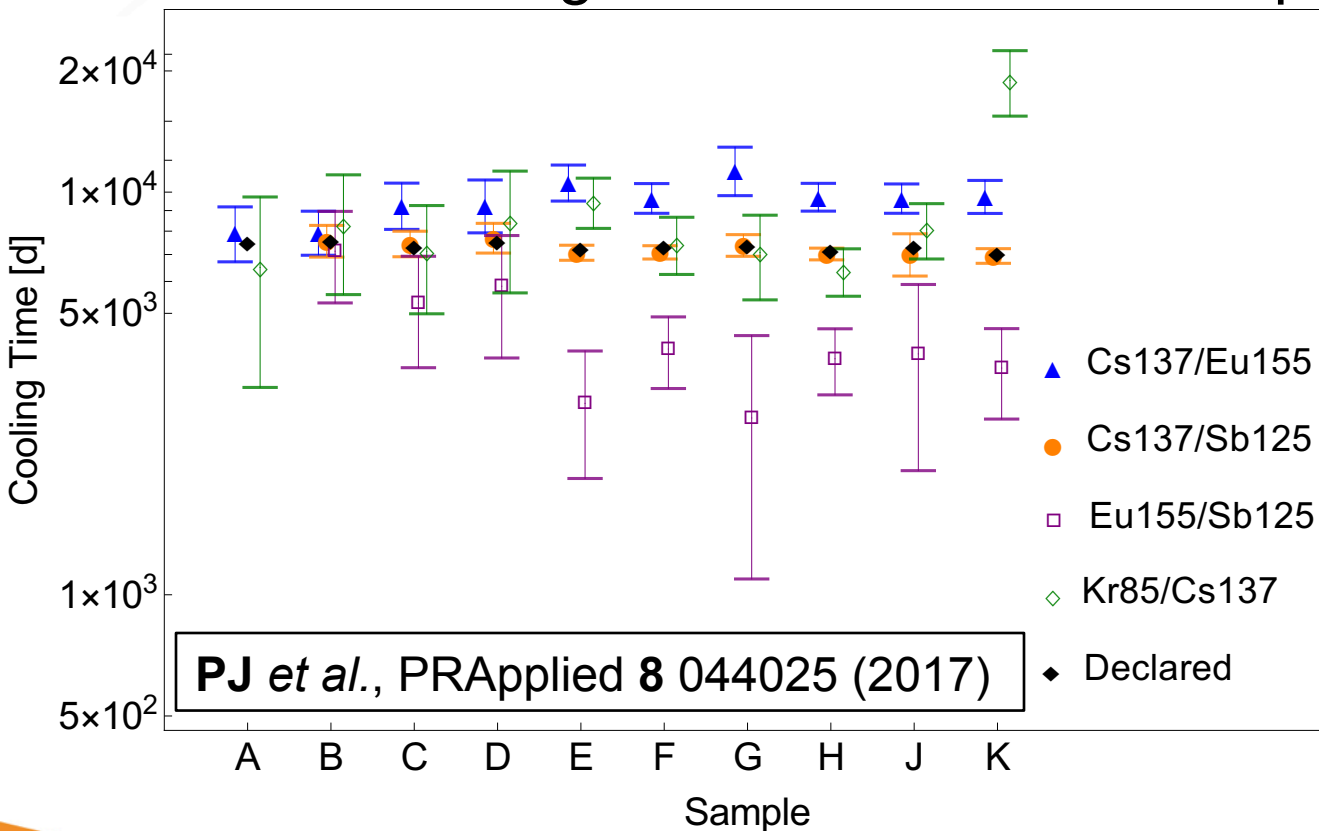
- Start with U-metal and UO_3 archived samples
 - Separate Pu and U then TIMS for isotopics
- Uranium isotopics indicate nat U for ϵ_0



UNCLASSIFIED

Applying the diagnostics

- Gamma spectrometry to identify fission products
 - Measured ^{85}Kr , ^{125}Sb , ^{137}Cs , ^{152}Eu , ^{154}Eu , ^{155}Eu
 - Perform diagnostics on linear fission products



- Multiple T_C diagnostics required!
- $^{137}\text{Cs}/^{125}\text{Sb}$ matches declared T_C

UNCLASSIFIED

Conclusion

- Fission is an extremely complex and intricate process
 - Tremendous modeling progress made so far
 - Stepping towards a predictive fission model: $Y_{\text{pre}}(A) + \text{CGMF}$
- Applications are far-reaching
 - Reactor heating, nonproliferation, forensics, fund. science
- Tools and models exist to fill in the gaps where experiments cannot or have not been yet
 - Ability to improve accuracy of applications
 - Find new applications and new designs

UNCLASSIFIED

Next Steps

1. Fine-tune and perform optimization on Pu suite
 - Find $\langle \text{TKE} \rangle$ and $d\langle \text{TKE} \rangle / dE_n$ that produce reasonable neutron properties
 - Include calculations of spontaneous fission
2. Expand the U suite as well
 - Also useful for criticality, reactor heating, etc.
3. Begin evaluation procedure to get consistent fission data for ^{235}U , ^{238}U , ^{239}Pu
 - Identified as high-priority for nuclear data community

UNCLASSIFIED

Thank you for your attention!

References & Funding

- **PJ** and P. Huber PRL **116** 122503 (2016)
- A. Chyzh, **PJ**, *et al.* (submitted to PLB) (2017)
- **PJ** *et al.*, PRApplied **8** 044025 (2017)
- **PJ**, arXiv:1709.01183 (accepted in NSE 2018)
- **PJ**, P. Möller, A Sierk, P. Talou arXiv:1712.05511 (accepted in PRC 2018)
- S. Okumura, T. Kawano, P. Talou, **PJ**, S. Chiba arXiv:1802.01248 (2018)
- N. Fotiadis, **PJ**, *et al.* (in preparation) (2018)

U.S. Department of Energy (DOE)
National Nuclear Security Administration (NA-22)

UNCLASSIFIED

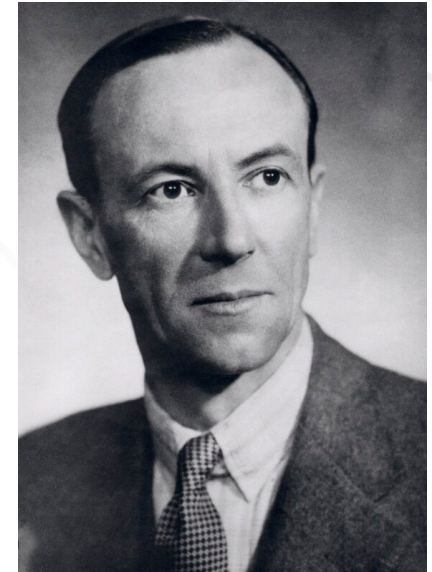
Extra



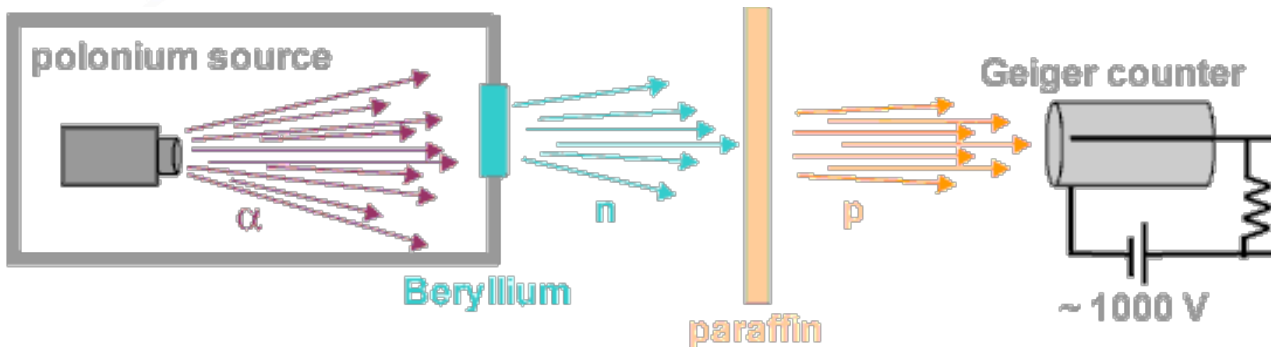
UNCLASSIFIED

History of nuclear fission

- Gateway to Fission
 - 1932: Chadwick discovers the neutron



James Chadwick



Possible Existence of a Neutron

James Chadwick

Nature, p. 312 (Feb. 27, 1932)

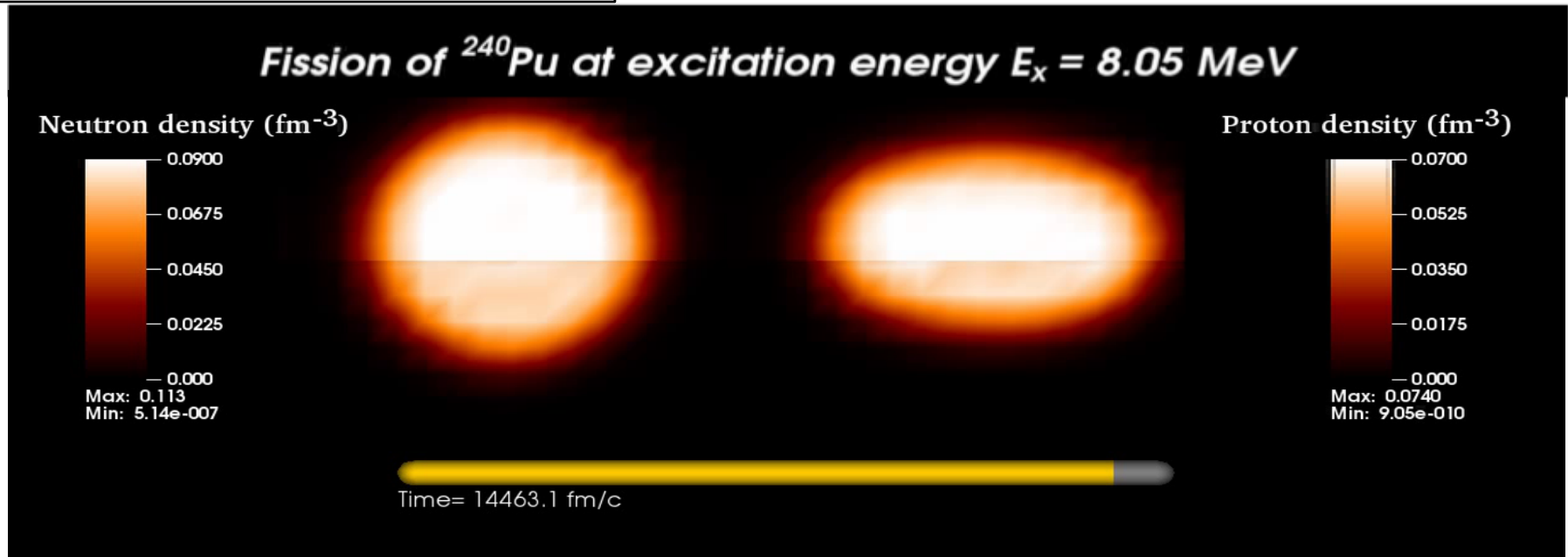
These results, and others I have obtained in the course of the work, are very difficult to explain on the assumption that the radiation from beryllium is a quantum radiation, if energy and momentum are to be conserved in the collisions. The difficulties disappear, however, if it be assumed that the radiation consists of particles of mass 1 and charge 0, or neutrons. The capture of the α -particle by the Be^9 nucleus may be supposed to result in the formation of a C^{12}

History of Nuclear Fission

6. Nuclear Evolution (today)

- Micro: based on nucleon-nucleon forces (calculate densities)
- Computationally expensive (not ideal for yields)

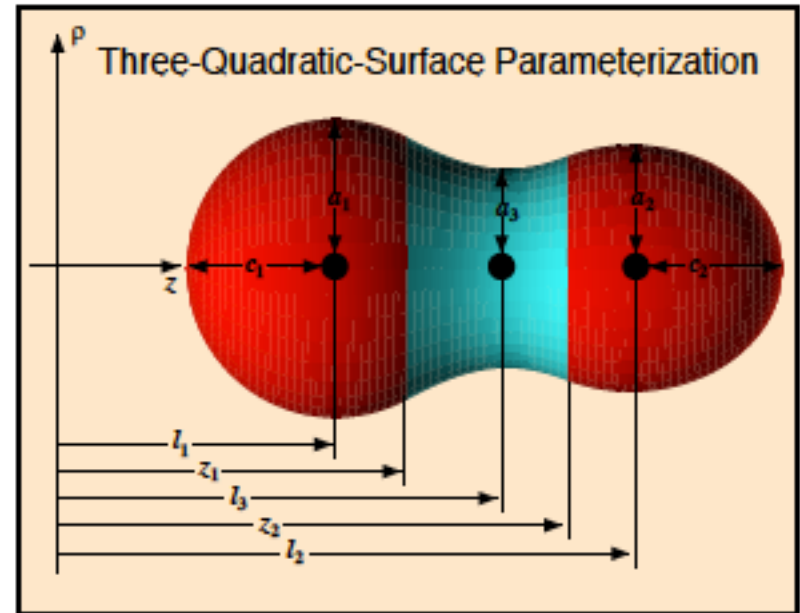
Bulgac, PRL **116** 122504 (2016)



UNCLASSIFIED

Macro-micro fission yields

- Compute the potential energy surface of a fissioning nucleus
 - Macroscopic shape + microscopic shell/pairing corrections
 - Macroscopic shape given by collection of shape variables q_i
 - 5D for $Y_{\text{pre}}(A)$ and 6D for $Y_{\text{pre}}(A, Z)$



P. Möller & T. Ichikawa EPJ A **51** 173 (2015)

Nuclear shape

$$E_T(Z, N, q_i) = E_M(Z, N, q_i) + E_m^p(Z, N, q_i) + E_m^n(Z, N, q_i) + E_{\text{odd}}$$

Loose dependence on N/Z

Nucleon corrections

UNCLASSIFIED

Hauser-Feshbach Model

$$P(\epsilon_\gamma)dE \propto T_\gamma(\epsilon_\gamma)\rho(Z, A, E - \epsilon_\gamma)dE$$

$$P(\epsilon_n)dE \propto T_n(\epsilon_n)\rho(Z, A - 1, E - \epsilon_n - S_n)dE$$

- Need transmission coefficients and level densities

$T_\gamma(\epsilon_\gamma)$ from strength-function formalism

$$T^{XL}(\epsilon_\gamma) = 2\pi f_{XL}(\epsilon_\gamma)\epsilon_\gamma^{2L+1}$$

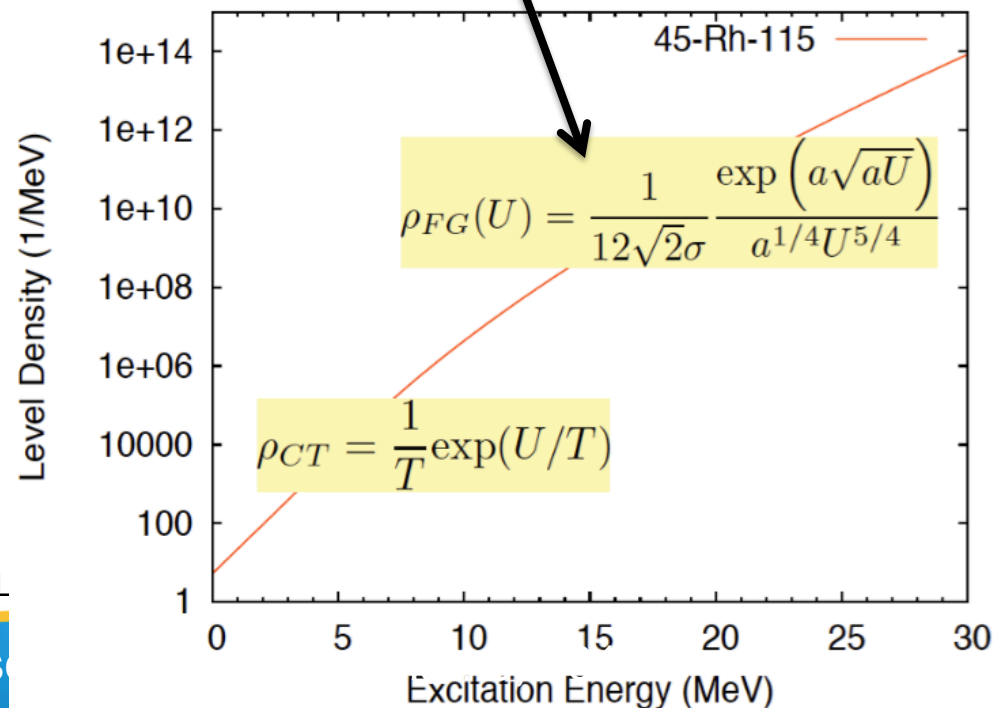
$T_n(\epsilon_n)$ from optical model calculations

$$T_c = 1 - \left| \langle S_{cc} \rangle \right|^2$$

RIPL discrete levels

+

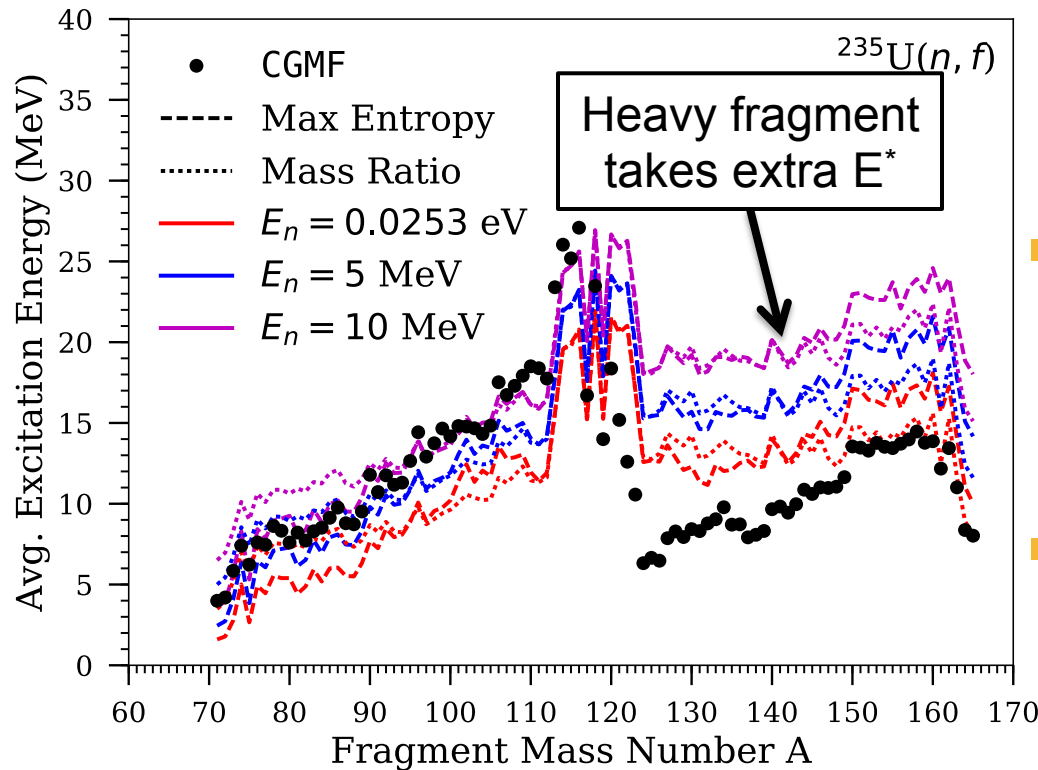
Gilbert-Cameron with $a(U)$



UNCL

Excitation Energy Sharing

- Currently: vary R_T until $v(A)$ matches (CGMF)



$$\frac{E_L^*}{E_H^*} = \frac{a_L}{a_H} R_T^2 \longrightarrow E_H^* = \frac{E_T^* a_H}{a_L R_T^2 + a_H}$$

- Next: share E_{int} via maximum entropy
- Similar structure as fitted $R_T(A)$

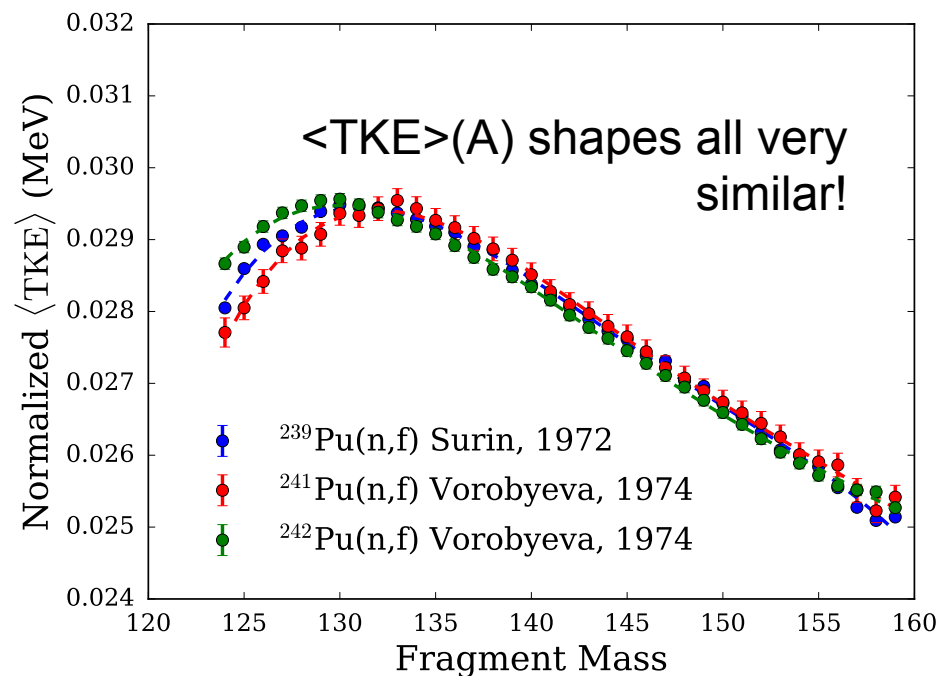
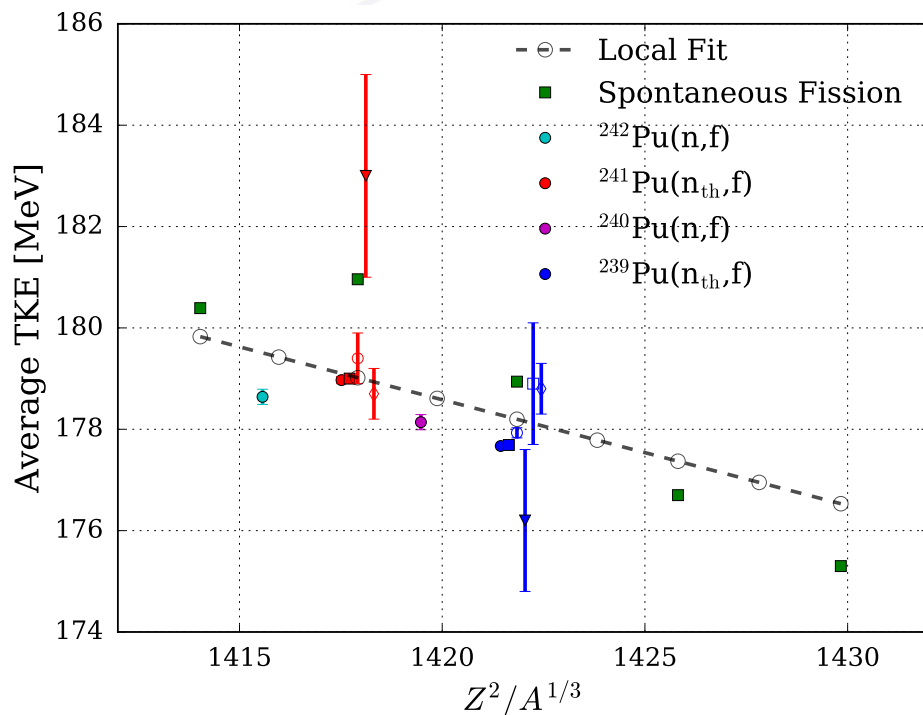
Max Entropy

$$\langle E_H \rangle = \frac{\int_0^{E_{\text{int}}} \varepsilon \rho_H(\varepsilon) \rho_L(E_{\text{int}} - \varepsilon) d\varepsilon}{\int_0^{E_{\text{int}}} \rho_H(\varepsilon) \rho_L(E_{\text{int}} - \varepsilon) d\varepsilon}$$

UNCLASSIFIED

<TKE> systematics

- <TKE> is from Coulomb-repulsion $\rightarrow Z^2/A^{1/3}$ form



- <TKE>(A) shapes from nearby Pu
- J^π distribution is a Gaussian

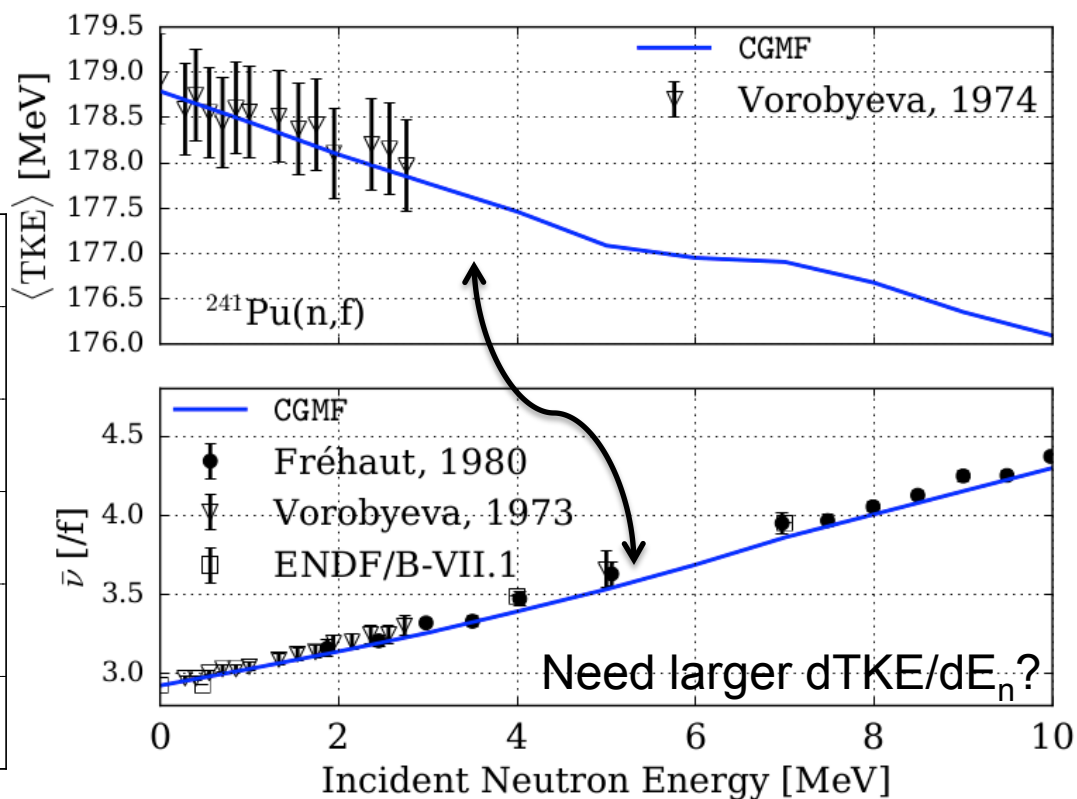
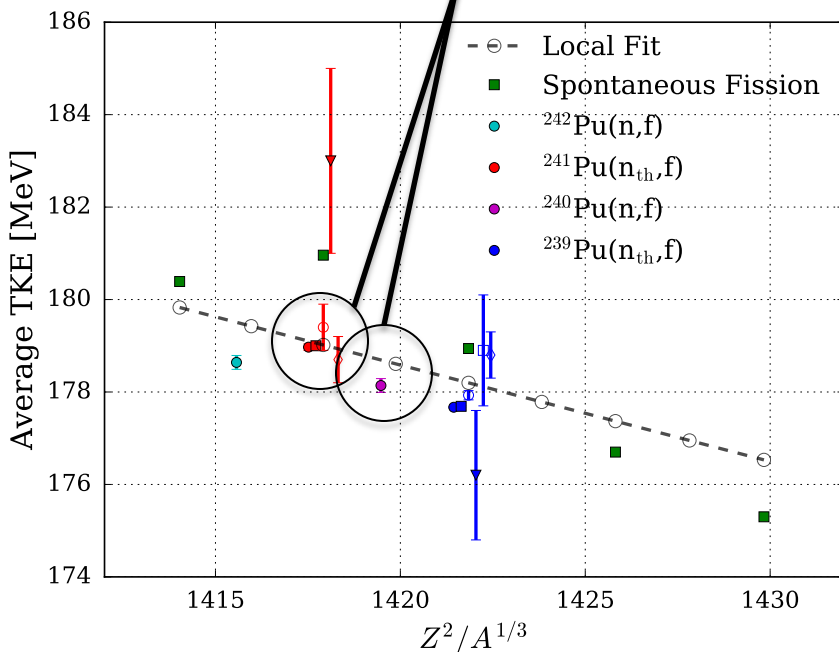
Becker, PRC **87** 014617 (2013)

UNCLASSIFIED

Neutron- $\langle TKE \rangle$ correlation results

- More $\langle TKE \rangle$, less $\langle TXE \rangle$ and fewer neutrons!
- Validate** systematics with known Pu isotopes!
- Can use avg. n multiplicity to place constraints on $\langle TKE \rangle$

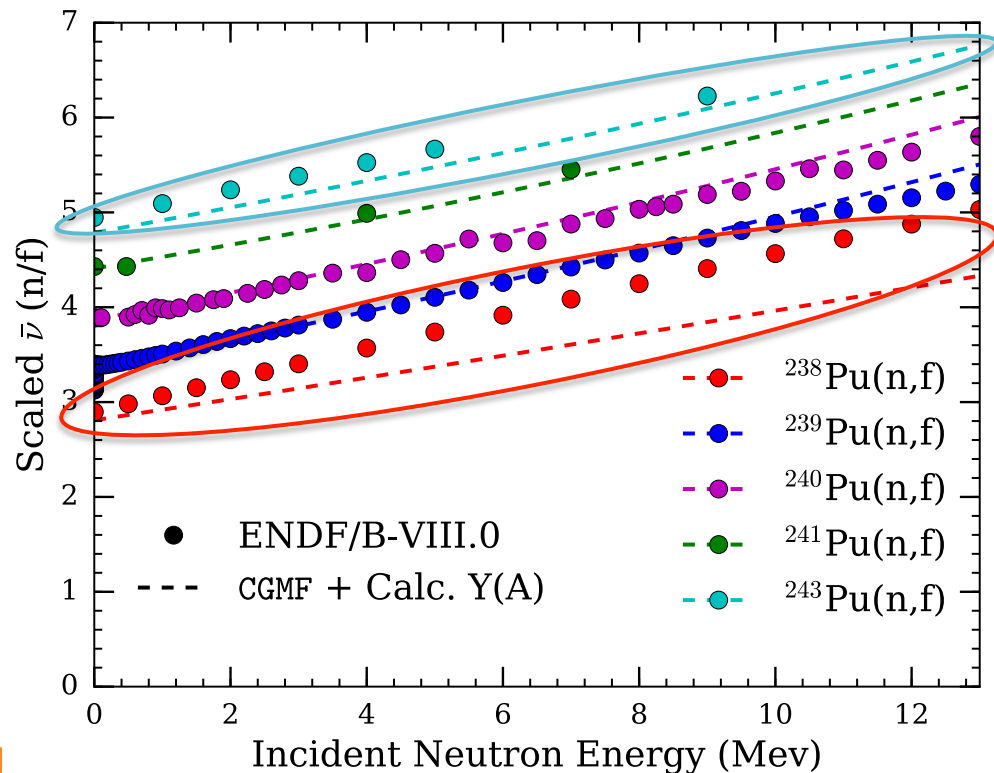
Pu241 TKE linked to Pu242 TKE via 2nd chance!



UNCLASSIFIED

Predictions for neutron characteristics

- Prompt neutron multiplicity agrees with ENDF/B-VIII
 - Shift differences could indicate $\langle TKE \rangle$ inaccuracy
 - Slope differences could be from bad $d\langle TKE \rangle/dE_n$

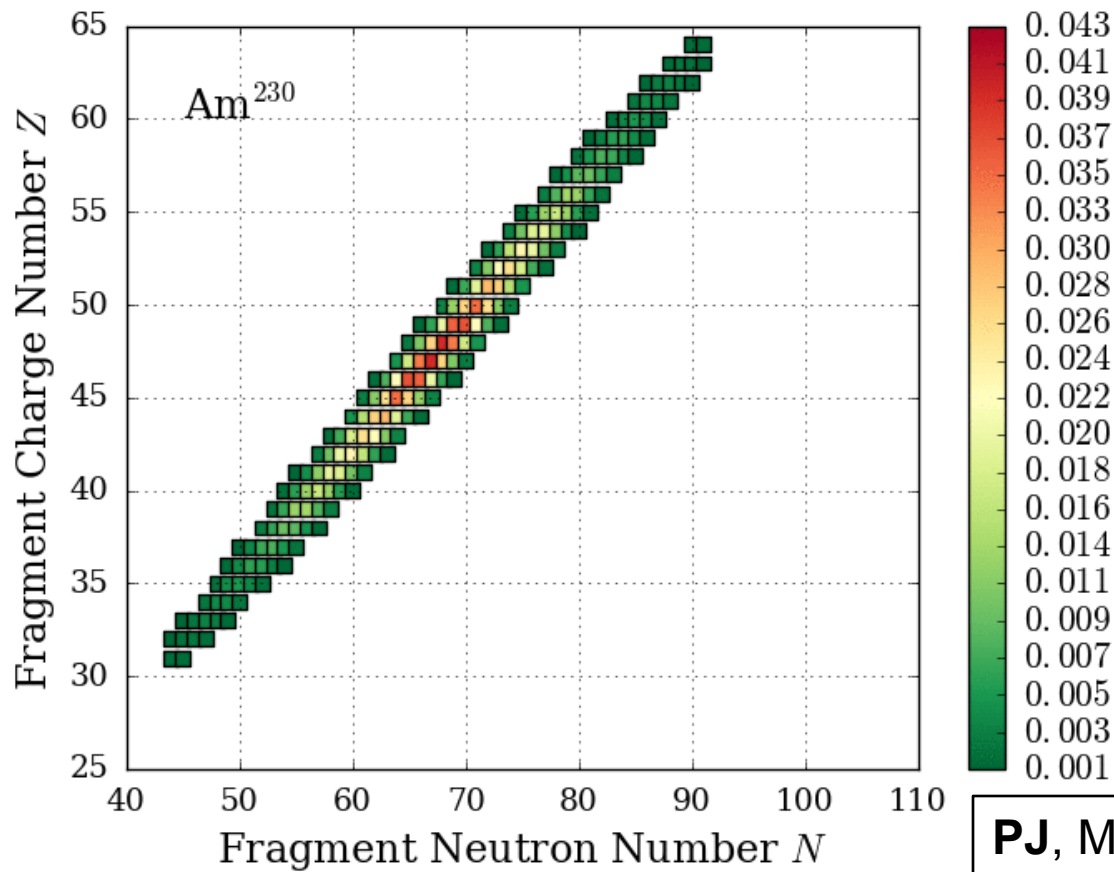


We must be careful not to take ENDF as nature! Can often be evaluations/predictions another physicist performed!

FIED

Application – fission in r-process (FIRE)

- Developing $Y_{\text{pre}}(A, Z)$ systematics for r-process
 - Use macro-micro $Y_{\text{pre}}(A)$ and apply simple $Y(Z|A)$

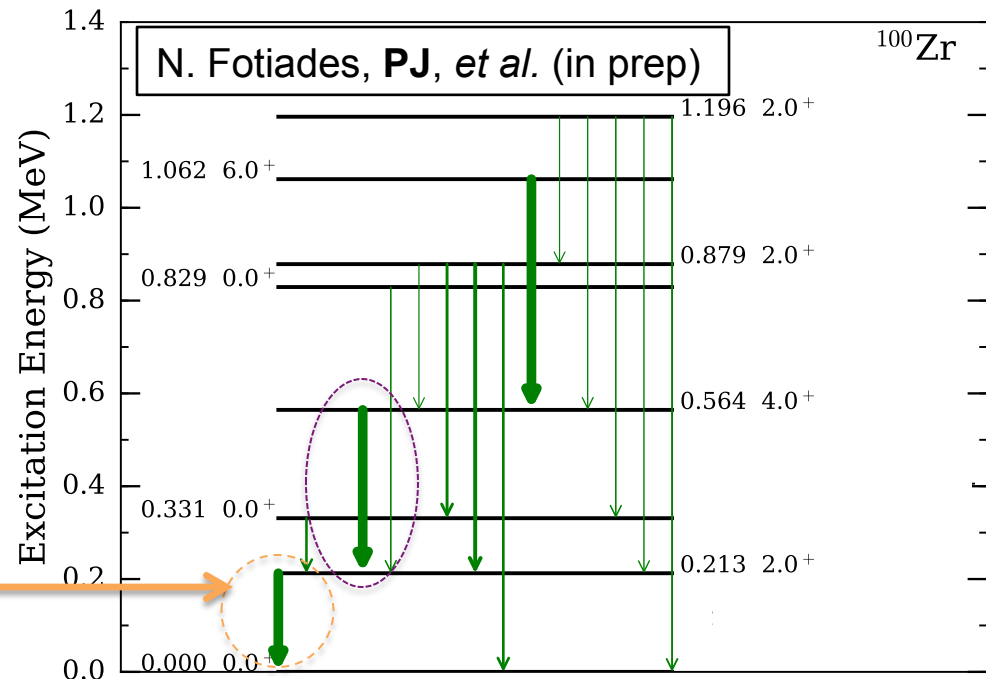
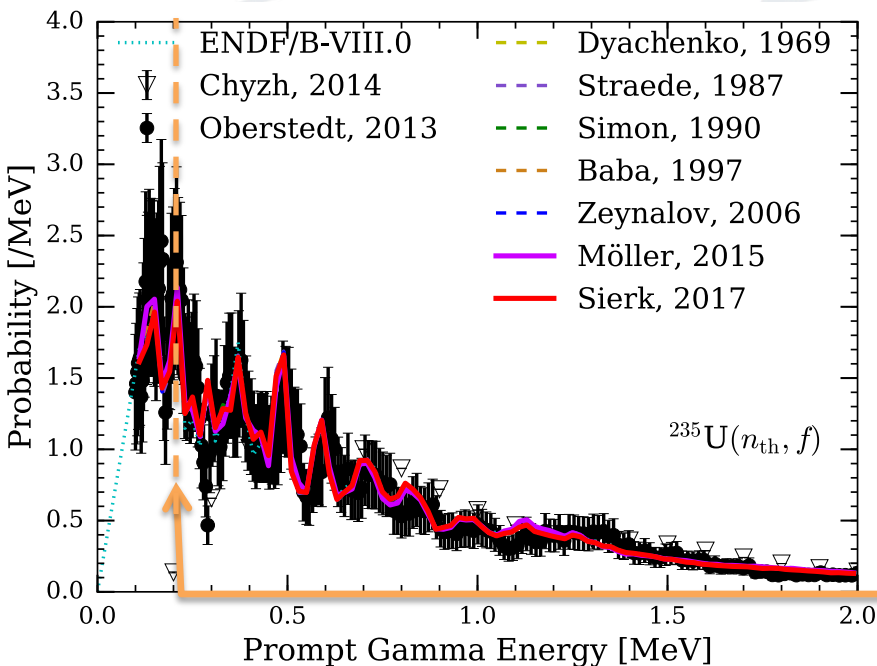


- Americium isotopes show transition from symmetric to asymmetric
- May need to include sf, βf , etc.

PJ, M. Mumpower, P. Möller (in prep)

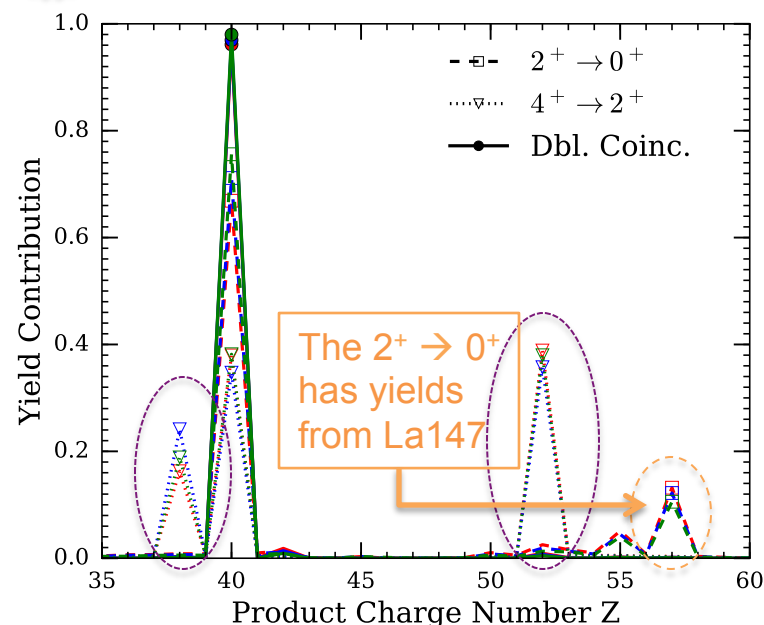
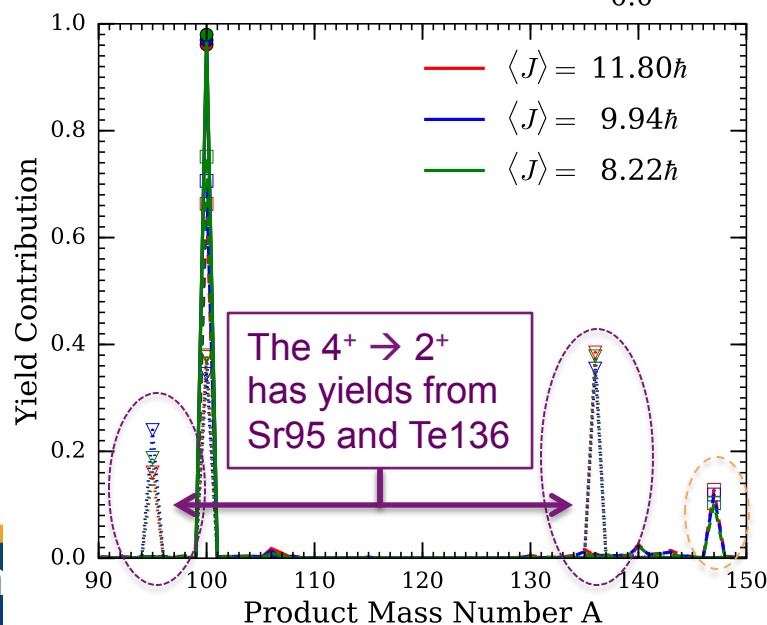
UNCLASSIFIED

3) Gamma-ray spectroscopy



Analyze γ -ray spec data and infer the yields of products with CGMF!

Double-gating on both the $4^+ \rightarrow 2^+$ and the $2^+ \rightarrow 0^+$ is almost 98% clean!



Developing diagnostics for very-low burnup

■ Neutron Exposure: Uranium ratios

$$\Phi_n = \frac{\ln(\varepsilon_0 / \varepsilon)}{\sigma_{U235}^T - \sigma_{U238}^T}$$

$$\varepsilon = {}^{235}\text{U}/{}^{238}\text{U}$$

$$\Phi_n = \frac{1}{\sigma_{U235}^T - \sigma_{U236}^T} \ln \left(\frac{\sigma_{U235}^C - \rho(\sigma_{U236}^T - \sigma_{U235}^T)}{\sigma_{U235}^C} \right)$$

$$\rho = {}^{236}\text{U}/{}^{235}\text{U}$$

■ ${}^{235}\text{U}/{}^{238}\text{U}$:

- Relies on knowing initial enrichment ✗
- More accurate as concentration is higher ✓

■ ${}^{236}\text{U}/{}^{235}\text{U}$:

- Trouble when ${}^{236}\text{U}$ is very low ✗
- Independent of initial enrichment ✓

UNCLASSIFIED

Deriving the cooling time diagnostic

- Linear Systems: Simplest reaction networks

$$\frac{dN_L}{dt} = -(\lambda_L + \phi_n \sigma^T) N_L + \vec{Z}_L \cdot \vec{F}$$

Depleted via β -decay and n-capture
Produced via fission

- For most linear nuclides, β -decay dominates...

$$\frac{dN_L}{dt} = -(\lambda_L + \cancel{\phi_n \sigma^T}) N_L + \vec{Z}_L \cdot \vec{F} \xrightarrow{\text{Solution}} N_L(t) = \frac{\vec{Z}_L \cdot \vec{F}}{\tilde{\lambda}_L} (1 - e^{-\tilde{\lambda}_L t})$$

- Taylor expand...

Assumes no initial abundance at start of irradiation \rightarrow satisfied for linear systems

$$N_L(t) = \frac{\vec{Z}_L \cdot \vec{F}}{\tilde{\lambda}_L} (1 - [1 - \tilde{\lambda}_L t + \frac{1}{2} (\tilde{\lambda}_L t)^2 \dots])$$

Add in $e^{-\lambda^T}$ for decay time

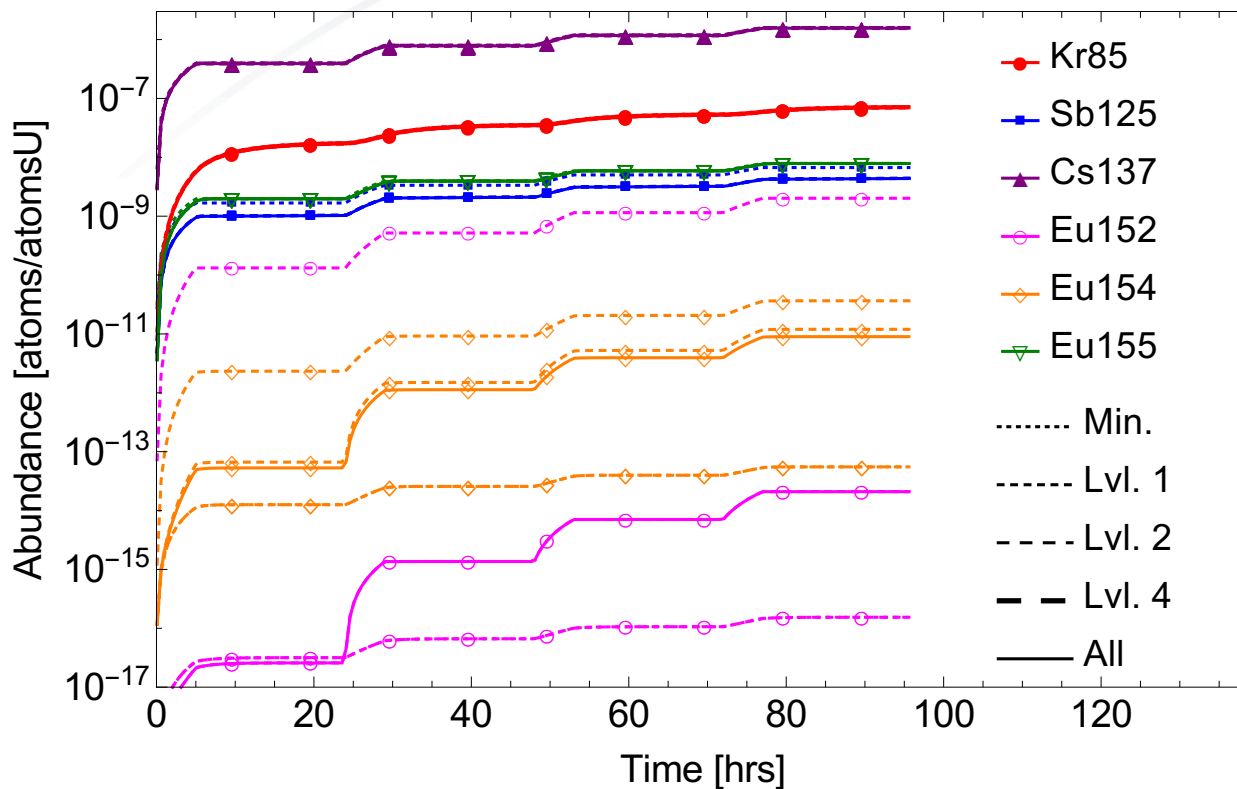
$$\hookrightarrow N_L(t) = t(\vec{Z}_L \cdot \vec{F}) \longrightarrow \Phi_n \left(\vec{Z}_L \cdot \langle \vec{\Sigma}_{fiss} \rangle \right) e^{-\lambda_L T_c}$$

UNCLASSIFIED

Defining linear systems

Linear Systems:

PJ et al., PRApplied 8 044025 (2017)



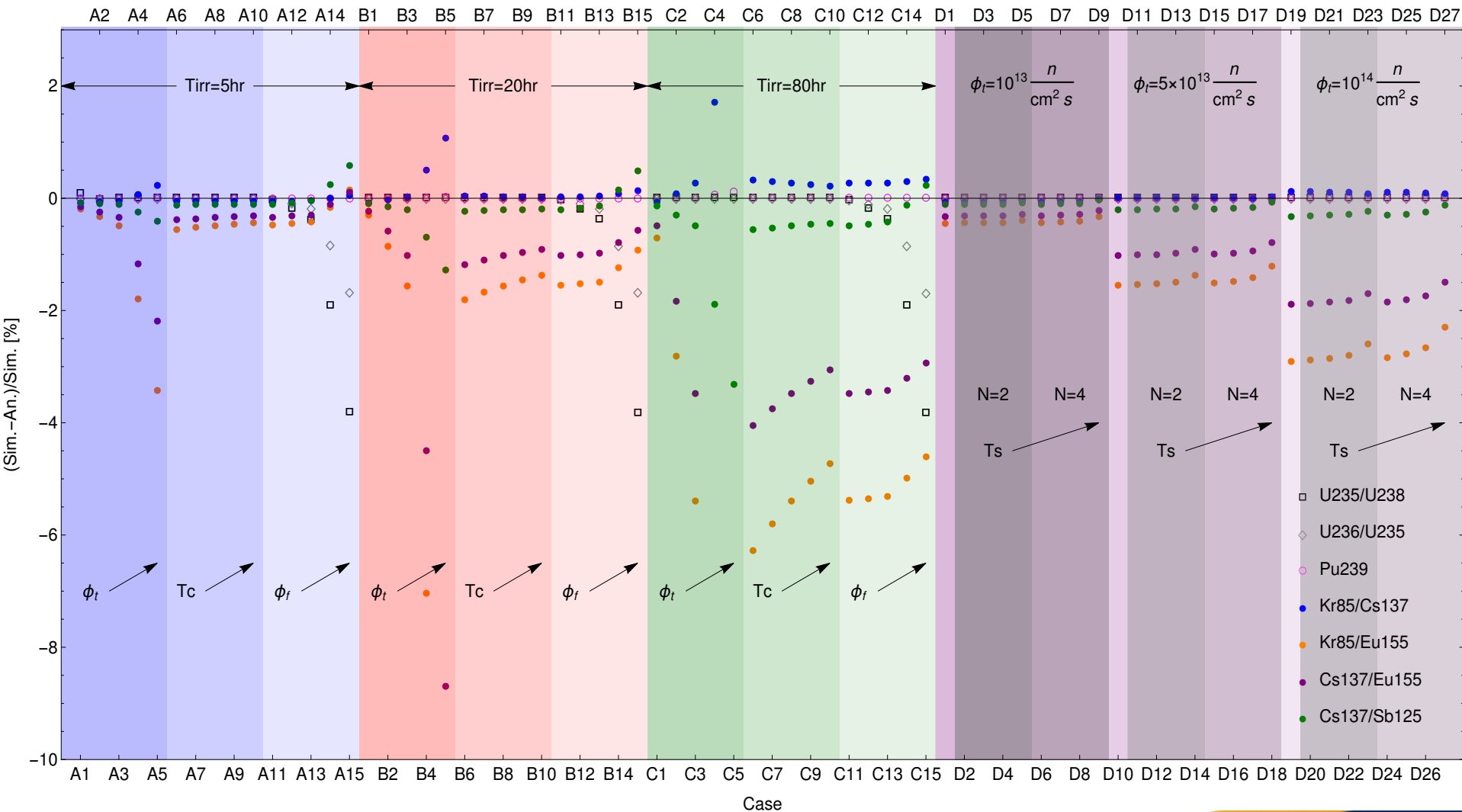
1. β -parents are short-lived
2. No significant neutron-capture channels
3. Large cumulative yields (for measurement purposes)
4. Long-lived (for measurements purposes)

Nonlinear depend on size of reaction network! ($^{152,154}\text{Eu}$ for example)

PJ and P. Huber PRL 116 122503 (2016)

UNCLASSIFIED

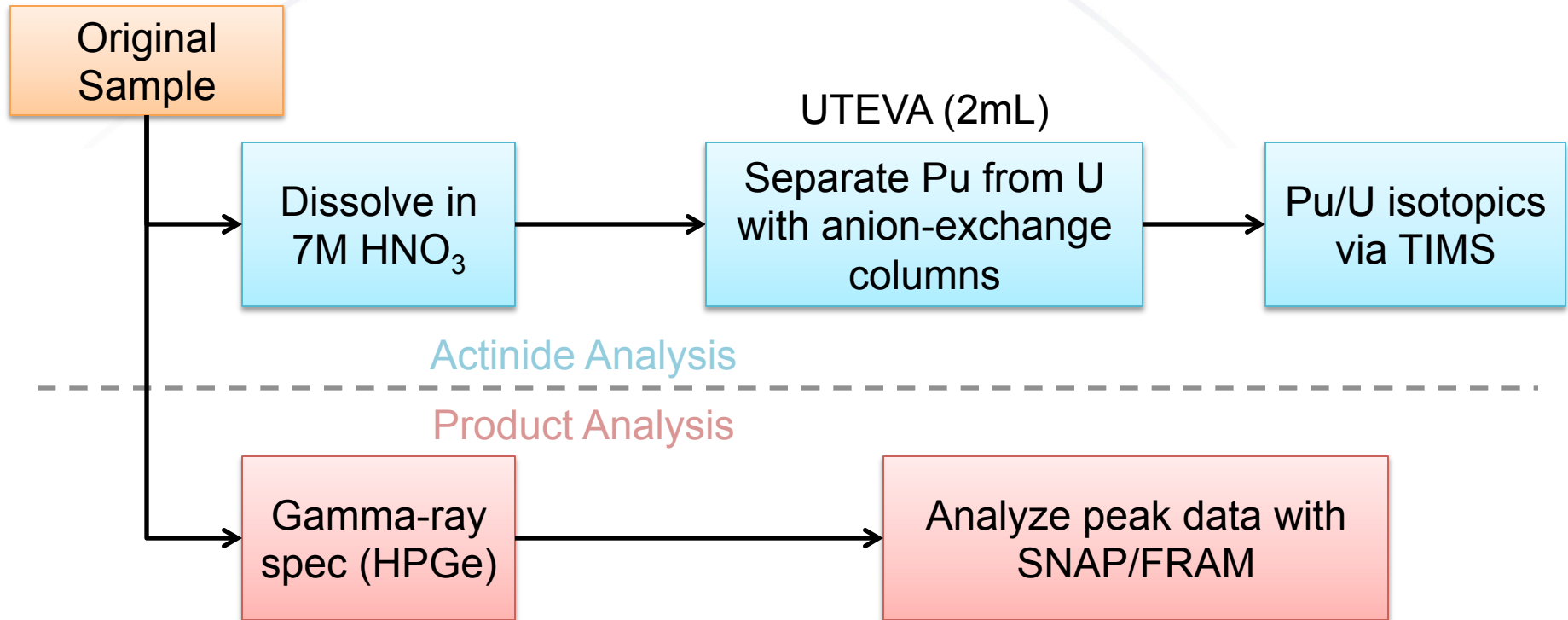
Verifying the diagnostics



UNCLASSIFIED

Chemical separation

- Start with U-metal and UO_3 archived samples



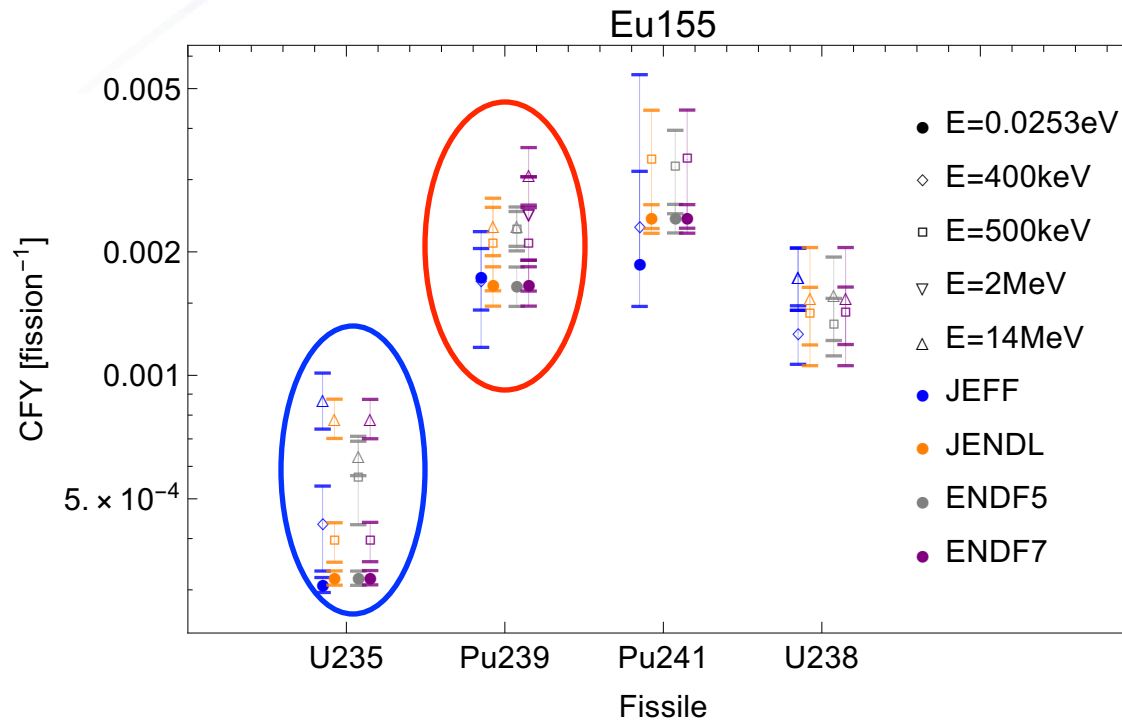
B. Byerly *et al.*, J. Radioanal Nucl Chem **307** (2016)

L. Tandon *et al.*, J. Radioanal Nucl Chem **282** (2009)

UNCLASSIFIED

Why flux-averaging is needed

- Fission product yields change with fissioning isotopes and with energy (and data source!)



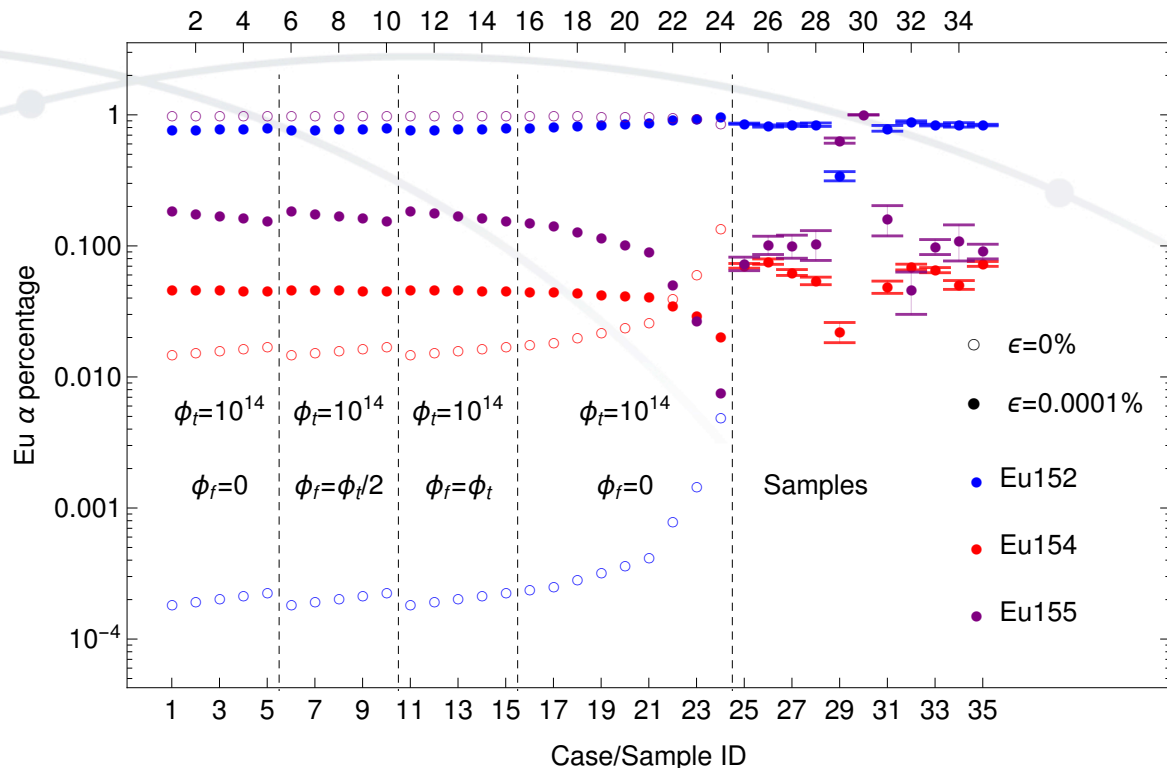
- **Pu239** yields are ~4x larger than **U235** yields!
- All databases are very similar

- Flux-average the fission rates so as not to bias towards U235 or Pu239 fissions

UNCLASSIFIED

Eu155 issue

- T_c estimates using Eu155 show systematic disagreement with others
- Abnormally large **Eu152**, **Eu154** abundances
- nat Eu?



Case/Sample ID					
151Eu ≥1.7E+18 Y 47.81% α	152Eu 13.517 Y ε: 72.10% β-: 27.90%	153Eu STABLE 52.19%	154Eu 8.601 Y β-: 99.98% ε: 0.02%	155Eu 4.753 Y β-: 100.00%	156Eu 15.19 D β-: 100.00%
150Sm STABLE 7.38%	151Sm 90 Y β-: 100.00%	152Sm STABLE 26.75%	153Sm 46.284 H β-: 100.00%	154Sm STABLE 22.75%	155Sm 22.3 M β-: 100.00%
149Pm 53.08 H β-: 100.00%	150Pm 2.698 H β-: 100.00%	151Pm 28.40 H β-: 100.00%	152Pm 4.12 M β-: 100.00%	153Pm 5.25 M β-: 100.00%	154Pm 2.68 M β-: 100.00%